OPTICAL RECORDING OF SHEAR FLOW OF A NEMATIC LIQUID CRYSTAL IN A GASDYNAMIC FLOW

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The theoretical and experimental study of the shear flow of non-Newtonian structured liquids, which include nematic liquid crystals (NLCs), is of scientific interest in itself and has practical potential.

As mentioned in [1], the use of liquid crystals in aerodynamic experiments can lead to new diagnostical methods, which in a number of cases are more informative and simpler than other known methods, In [1] we considered the case when a thin NLC layer was deposited on the surface of a model with gas flowing over it. The shear flow occurring in the NLC and the corresponding reorientation of the long axes of the crystal molecules (the direction \mathbf{n}) were computed numerically. From the data we found the optical response of the entire system. In the resulting algorithm such characteristics as the velocity of the gas flow on the free surface of the NLC and the shear stress constant are calculated from the change in the phase of the extraordinary light wave as it traverses the layer.

One of the difficulties in the practical implementation of the method proposed in [1] is that it requires a thin layer of NLC with a homeotropic (normal) orientation of the director on a solid substrate and with a free upper boundary, since the crystal forms in drops. It is of interest, therefore, to consider the case of a planar (tangential) geometry, for which **n** on the surface $(\mathbf{n}(z = 0) = \mathbf{n}_0)$ is orthogonal to both the direction of gas flow and the velocity gradient (Fig. 1; this configuration may be disrupted in the bulk $\mathbf{n}(z \neq 0)$).

The shear flow of a NLC in the geometry given above is studied here. The difference between the problem solved in [1] from that formulated here is that the deformation of the director no longer takes place in one plane. As a result, two angles must be considered for the description of the reorientation on the NLC molecules: the polar angle θ and the azimuthal angle φ (Fig. 1). Moreover, as becomes clear below, if for any reason the director cannot emerge form the x, y plane (th = 0), then reorientation with respect to φ does not appear either ($\varphi = 0$).

The vector **n** is expressed in terms of θ and φ as (Fig. 1).

$$\mathbf{n} = \{n_x, n_y, n_z\} = \{\cos\theta\cos\varphi, \ \cos\theta\sin\varphi, \ \sin\theta\}; \tag{1}$$

the velocity vector has one nonzero component

$$\mathbf{v} = \{0, o_{u}, 0\}.$$



UDC 532.783



Since the problem is one-dimensional, we assume that all the desired functions depend only on the time t and one spatial variable z. The velocity gradient w has one component

$$\mathbf{w} = \{0, 0, \frac{\partial v_y}{\partial z}\}, \ w_z = \frac{\partial v_y}{\partial z} \equiv w.$$

The equations necessary for describing the reorientation of the director by the shear flow are obtained from the Erickson-Leslie equations [2], which are written in general form as

$$\rho\left(\frac{\partial v_i}{\partial t} + v_j\frac{\partial v_i}{\partial x_j}\right) = f_i + \frac{\partial \Sigma_{ji}}{\partial x_j}, \ i,j = x,y,z.$$
(2)

Here ρ is the density of the liquid; f is the volume density of the forces acting on the NLC (in our case $\mathbf{f} = -\nabla \mathbf{p}$ (p is the pressure) and below we consider the case with $\nabla \mathbf{p} = 0$ and, hence, $\mathbf{f} = 0$); the viscous tensor is

$$\Sigma_{ij} = \alpha_1 n_i n_j A_{km} n_k n_m + \alpha_2 n_i N_j + \alpha_3 n_j N_i + \alpha_4 A_{ij} + \alpha_5 n_i n_k A_{kj} + \alpha_6 A_{ik} n_k n_j;$$

 α_i are the viscous parameters of the NLC; and

$$N_{i} = \frac{dn_{i}}{dt} + n_{j}\omega_{ji}; A_{ij} = \frac{1}{2} \left(\frac{\partial v_{i}}{\partial x_{j}} + \frac{\partial v_{j}}{\partial x_{i}} \right);$$
$$\omega_{ij} = \frac{1}{2} \left(\frac{\partial v_{i}}{\partial x_{j}} - \frac{\partial v_{j}}{\partial x_{i}} \right).$$

Equations describing the motion of the director must be added to (2) in order to close the system. Ignoring the inertial terms, in accordance with the variational principle [2] we obtain

$$\frac{\partial}{\partial x_i} \frac{\delta \mathcal{F}}{\delta (\partial n_i / \partial x_i)} - \frac{\delta \mathcal{F}}{\delta n_j} = [\mathbf{n}, \mathbf{R}]_j + \lambda_L n_j, \qquad (3)$$

where λ_L is an indeterminate Lagrangian multiplier, which ensures satisfaction of the condition that **n** is unique; $|\mathbf{n}| = (n_x^2 + n_y^2 + n_2^2)^{1/2} = 1$; and \mathcal{F} is the density of free energy. In the absence of external fields \mathcal{F} has the form

$$\mathcal{F} = \frac{1}{2} \{ K_1(\operatorname{div} n)^2 + K_2(n, \operatorname{rot} n)^2 + K_3(n, \operatorname{rot} n)^2 \}.$$

Here R is the dissipative force expressed in terms of the vector N and the symmetric part A_{ii} of the velocity-gradient tensor.

$$R_i = \gamma_1 N_i + \gamma_2 n_j A_{ji}$$

($\gamma_1 = \alpha_3 - \alpha_2, \gamma_2 = \alpha_6 - \alpha_5$).

All of the relations are simplified in the given case of the one-dimensional shear flow of a NLC, when the desired functions depend only on one coordinate z. Equation (2) thus becomes

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$$\rho \frac{\partial v_y}{\partial t} = \frac{\partial \Sigma_{zy}}{\partial z}.$$
 (4)

Only one component is needed:

$$\Sigma_{zy} = \alpha_1 n_z n_y w n_z n_y + \alpha_2 n_z \left(\frac{\partial n_y}{\partial t} - n_x \frac{w}{2} \right) + \alpha_3 n_y \left(\frac{\partial n_z}{\partial t} + n_y \frac{w}{2} \right) + \alpha_4 \frac{w}{2} + \alpha_5 n_z \frac{w}{2} + \alpha_6 n_y \frac{w}{2}.$$
(5)

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The values of ω_{ij} and A_{ij} , are written as a matrix with two nonzero elements,

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$$\omega_{ij} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & \frac{w}{2} \\ 0 & -\frac{w}{2} & 0 \end{pmatrix}, A_{ij} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & \frac{w}{2} \\ 0 & \frac{w}{2} & 0 \end{pmatrix},$$
(6)

and the vectors N and R are written as

$$\mathbf{N} = \left\{ \frac{\partial n_x}{\partial t}, \frac{\partial n_y}{\partial t} - n_z \frac{w}{2}, \frac{\partial n_z}{\partial t} + n_y \frac{w}{2} \right\},$$
$$\mathbf{R} = \left\{ \gamma_1 \frac{\partial n_x}{\partial t}, \gamma_1 \left(\frac{\partial n_y}{\partial t} - n_z \frac{w}{2} \right) + \gamma_2 \frac{w}{2} n_z, \gamma_1 \left(\frac{\partial n_z}{\partial t} + n_y \frac{w}{2} \right) + \gamma_2 n_y \frac{w}{2} \right\}.$$
(7)

The equation for the density of free energy $\mathcal F$ also simplifies to

$$\mathcal{F} = \frac{1}{2} \left\{ K_1 \left(\frac{\partial n_z}{\partial z} \right)^2 + K_2 \left(n_x \frac{\partial n_y}{\partial z} - n_y \frac{\partial n_z}{\partial z} \right)^2 + K_3 \left[\left(\frac{\partial n_x}{\partial z} \right)^2 + \left(\frac{\partial n_y}{\partial z} \right)^2 + \left(\frac{\partial n_z}{\partial z} \right)^2 \right] \right\}.$$
(8)

Only one term must be left in the variational relation (3):

$$\frac{\partial}{\partial z} \frac{\delta \mathcal{F}}{\delta(\partial n_j/\partial z)} - \frac{\delta \mathcal{F}}{\delta n_j} = [\mathbf{n}, \mathbf{R}]_j + \lambda_L n_j.$$
(9)

On substituting all the necessary quantities (5)-(8) into (4) and (9) and going over to angular notation of the vector **n** (1), we obtain the desired system of equations, which describe the shear flow of a NLC layer:

$$\gamma_{1}\frac{\partial\theta}{\partial t} = F(\theta)\frac{\partial^{2}\theta}{\partial z^{2}} + \frac{1}{2}\frac{\partial F(\theta)}{\partial \theta}\left(\frac{\partial\theta}{\partial z}\right)^{2} - \frac{1}{2}\frac{\partial G(\theta)}{\partial \theta}\left(\frac{\partial\varphi}{\partial z}\right)^{2} - \frac{1}{2}\frac{\partial \sigma}{\partial \theta}\left(\frac{\partial\varphi}{\partial z}\right)^{2} - \frac{1}{2}\frac{\partial\sigma}{\partial z}\left(\gamma_{1} + \gamma_{2}\cos 2\theta\right)\sin\varphi,$$

$$\gamma_{1}\cos^{2}\theta\frac{\partial\varphi}{\partial t} = \frac{\partial}{\partial z}\left[G(\theta)\frac{\partial\varphi}{\partial z}\right] - \frac{\alpha_{2}}{2}\frac{\partial\sigma}{\partial z}\sin 2\theta\cos\varphi,$$

$$\rho\frac{\partial\sigma}{\partial t} = \frac{\partial}{\partial z}\left[\frac{\partial\sigma}{\partial z}g(\theta,\varphi) + \frac{\partial\theta}{\partial t}g(\theta,\varphi) + \frac{\partial\varphi}{\partial t}p(\theta,\varphi)\right].$$
(10)

Here

$$F(\theta) = K_1 \cos^2 \theta + K_3 \sin^2 \theta,$$

$$G(\theta) = (K_2 \cos^2 \theta + K_3 \sin^2 \theta) \cos^2 \theta,$$

$$g(\theta,\varphi) = \frac{1}{2} \left[(2\alpha_1 \sin^2 \theta + \alpha_3 + \alpha_6) \cos^2 \theta \sin^2 \varphi + \alpha_4 \right]$$

$$+ (\alpha_5 - \alpha_2) \sin^2 \theta \right], \quad q(\theta,\varphi) = (\alpha_3 \cos^2 \theta - \alpha_2 \sin^2 \theta) \sin \varphi,$$

$$p(\theta,\varphi) = \frac{\alpha_2}{2} \sin 2\theta \cos \varphi.$$
(11)

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When **n** is written in the form (1) it is no longer necessary to introduce the Lagrangian multiplier λ_L into (9), since the normalization condition $|\mathbf{n}| = 1$ is satisfied automatically. From the second equation it follows that for $\theta = 0$ the azimuthal angle $\varphi = 0$, i.e., it does not "feel" the influence of the flow.

Even though it is simpler than the initial system, the system obtained is still fairly complicated for analytical study. Henceforth Eqs. (10) and (11) will be solved numerically, in much the same way as in [1].

How the NLC behaves under the given conditions can be understood if the system (10), (11) is simplified further and the analytical solution is found. For this purpose we make the following assumptions.

1. Let us consider the steady-state case. All the desired quantities are assumed to be independent of time: $\partial/\partial t = 0$.

2. The velocity gradient is nearly linear: w = const. In [1], when calculating the velocity profiles for the reorientation of a director from a homeotropic to a planar structure by a shear flow, we showed that the difference from linearity is small.

3. The flow is fairly strong and polar-angle saturation is attained $\theta = \theta_0$, where θ_0 is determined from the first equation of (10), if the elastic forces are ignored:

$$\cos 2\theta_0 = -\frac{\gamma_1}{\gamma_2}.$$

If we take into account the Parody relation [2] for the viscosity coefficients

$$\alpha_6 - \alpha_5 = \alpha_2 + \alpha_3,$$

we obtain

NLC.

$$tg^2\theta_0 = \frac{\alpha_3}{\alpha_2}.$$
 (12)

The angle θ_0 , as a rule, is small and is of the order of 10°.

4. The layer thickness L is assumed to be fairly large, greater than the characteristic changes of orientation λ in the

5. For simplicity we take the case of a single constant $K_1 = K$.

With the above assumptions the system of equations (10) is reduced to one equation for the azimuthal angle φ in the form

$$K\frac{d}{dz}\left(\cos\theta_{0}\frac{d\varphi}{dz}\right) - \frac{\alpha_{2}w\sin2\theta_{0}}{2}\cos\varphi = 0$$

or, taking Eq. (12) into account, we finally write

$$\frac{d^2\varphi}{dz^2} + \mu^2 \cos\varphi = 0, \tag{13}$$

where

$$\mu^2 = \frac{1}{\lambda^2} = \frac{\sqrt{\alpha_2 \alpha_3}}{K} w; \tag{14}$$

and λ is the characteristic dimension for which the main change of orientation occurs.

The expression for λ contains $(\alpha_2 \alpha_3)^{1/2}$. As a rule, the signs of α_2 and α_3 are negative [3] and steady-flow calculations pose no problems. In some types of NLC [3], however, $\alpha_3 > 0$ and (13) becomes meaningless in that case. It is known form experiments that crystals for which $\alpha_3 > 0$ [3] cannot undergo laminar flow at any velocity.

Equation (13) is solved analytically. To do this we multiply it by $d\varphi/dt$ and integrate and as a result the order of the equation decreases.

$$\frac{d\varphi}{dt} = \pm \sqrt{2} (\nu^2 - \mu^2 \sin\varphi)^{1/2}$$
(15)

(ν is a constant which is determined from the boundary conditions). Further integration leads to an elliptic integral. In our case we can use the condition that the characteristic changes of orientation in the crystal are small in comparison with the thickness L (L >> λ) and set the boundary condition as $z \rightarrow \infty$. In other words, far from the solid surface we expect to have $\varphi = \pi/2$ and $d\varphi/dz = 0$. The $\nu = \mu$ and, therefore, (15) becomes

$$\int \frac{d\varphi}{\left(1 - \sin\varphi\right)^{1/2}} = \pm \sqrt{2\mu}z + C_1$$

 $(C_1$ is a constant which is determined from the boundary condition).

After the final integration we have

$$\varphi = \frac{\pi}{2} - 4 \arctan\left[tg\frac{\pi}{8} \exp(\pm \mu z) \right].$$
 (16)

We have allowed for the fact that $\varphi = 0$ for z = 0. The \pm signs correspond to the rotation of NLC molecules to the right or left. For definiteness we take the minus sign. Note that $\tan \pi/8$ differs from $\pi/8$ by a mere 5%. Accordingly, if we do not require high accuracy, we can rewrite the solution (16) as

$$\varphi = \frac{\pi}{2} [1 - \exp(-\mu z)].$$
(17)

The flow causes reorientation of the director lying close to the xy plane with a characteristic transition-layer thickness $\lambda(w)$ (14).

We estimate the value of λ from the example of two NLCs, p-azoxyanizole (PAA) and N-(n-methoxybenzylidene)-nbutylaniline (MBBA). For PAA [3] $\alpha_2 \sim -0.066$ P, $\alpha_3 \sim -0.004$ P, and K ~ 5.6 $\cdot 10^{-7}$ dyne. The flow velocity gradient is w = 100 (cm/sec)/cm, whereby $\lambda_{PAA} \sim 6 \mu m$. For MBBA [3] $\alpha_2 \sim -0.78$ P, $\alpha_3 -0.01$ P, and K ~ 5.8 $\cdot 10^{-7}$ dyne, whereby $\lambda_{PAA} \sim 2.6 \mu m$.

It is of interest to study the variation of the apparent viscosity η_a as a function of the velocity gradient w. The following iteration step must be carried out in order to do this. In the steady-state case, the third equation of (10) gives

$$wg(\theta_0,\varphi) = b,$$

where b is the shear stress constant.

The apparent viscosity is determined from

$$\eta_a = bL/v_v(L). \tag{18}$$

In turn,

$$v_{y}(L) = \int_{0}^{L} w dz = b \int_{0}^{L} \frac{dz}{g(\theta_{0}, \varphi)}.$$
 (19)

Substituting (19) into (18), we obtain

$$\eta_a = L \begin{bmatrix} L \\ \int \frac{dz}{g(\theta_0, \varphi)} \\ 0 \end{bmatrix}^{-1}.$$
(20)

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If we assume that $\theta_0 \ll 1$, then Eq. (20) becomes

$$\eta_{a} = \frac{L}{2} \left[\int_{0}^{L} \frac{dz}{(\alpha_{3} + \alpha_{6})\sin^{2}\varphi + \alpha_{4}} \right]^{-1}.$$
(21)

When the velocity gradient (or the velocity of the upper boundary) is low, little reorientation occurs and, therefore $\varphi \approx 0$, whereupon it follows from (21) that $\eta_a \approx \alpha_4/2$. Conversely, for high velocity $\varphi \approx \pi/2$ and $\eta_a = (\alpha_3 + \alpha_4 + \alpha_6)/2$. The qualitative dependence $\eta_a(w)$, which follows from Eqs. (17) and (21), is shown in Fig. 2 (we do not give the calculation since it is not needed below).

The characteristic values of η_a for a PAA nematic liquid crystal are

$$\eta_{\mu}(w \rightarrow 0) \simeq 3.8 \text{ cP}, \eta_{\mu}(w \rightarrow \infty) \sim 1.9 \text{ cP}$$
 [3].

It is now necessary to consider the optical response of the given system to the shear action. The information obtained may consist of the phase and polarization when a beam of light is transmitted through the NLC, the polarization when a light beam is reflected from the free surface of the layer, or by combination of these methods. Compared to polarization changes, phase changes are technically more complicated since a reference beam is required but the pertinent calculations are generally simpler. Nevertheless these complexities can be eliminated, as is shown below.

When choosing the measuring procedure we must note that with a flow over the layer the liquid begins to leak, causing the layer thickness L to decrease, and L rather difficult to monitor. One way of overcoming this disadvantage was indicated in [1], but this requires measurements in two layers lying close together or use of a light ray reflected from the free surface.

The wave characteristics must be calculated on the basis of eh reduced Maswell's equations, since the geometricaloptics approximation used in [1] may be unsuitable for small values of λ and L.

A system of equations describing the propagation of a light wave through an arbitrarily oriented NLC sample is given in [4]. In our case, unlike that in [4], the transmitted wave is purely diagnostic, fairly low-power, and causes no additional distortions in the medium. For simplicity we assume that the transillumination is at a low angle to the surface of the later. The angle $\theta_0 \ll 1$, i.e., the nematic film is nearly planar in orientation throughout its volume. The equations from [4] then become

$$\frac{dA}{dz} = i \frac{\omega \varepsilon_a}{2cn_0} n(n,A), \qquad (22)$$

where the amplitude A(z) is related to the electric field E of the wave by $E = A(z)\exp(ikz-i\omega t)$ (ω is the frequency of the incident wave, $k = \omega/c$ is the wave vector, c is the speed of light in vacuum, n_0 is the refractive index for an ordinary wave, and ε_a is the dielectric anisotropy at the frequency ω). Since absorption is ignored, Eq. (22) has one integral (A, A^{*}) = const, which accords with the law of conservation of energy.

If we rewrite (22) by coordinates with allowance for (1) and introduce the dimensionless variable $\xi = z/L$, we obtain

$$\frac{dA_x}{d\xi} = iq'\cos\varphi(A_x\cos\varphi + A_y\sin\varphi),$$

$$\frac{dA_y}{d\xi} = iq'\sin\varphi(A_x\cos\varphi + A_y\sin\varphi),$$

$$0 \le \xi \le 1.$$
(23)

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For definiteness we take φ from (17).

$$\varphi = \frac{\pi}{2} [1 - \exp(-\mu'\xi)].$$

Here $q' = \varepsilon_a kL/2n_0$; $\mu' = \mu L$.

System (23) is rather complicated and its solution cannot be found analytically for an arbitrary value of $\varphi(z)$. Numerical computation is necessary here.

Before undertaking this computation, we recast the system (23) into a more convenient form. We redesignate the desired functions as

$$\Delta i = (A_{y}A_{y}^{*} - A_{x}A_{x}^{*})/I_{0},$$

$$i^{*} = (A_{x}A_{y}^{*} + A_{x}^{*}A_{y})/I_{0},$$

$$i^{-} = i(A_{x}A_{y}^{*} - A_{x}^{*}A_{y})/I_{0},$$
(24)

where

$$A_{v}A_{v}^{*} + A_{v}A_{v}^{*} = I_{0} = \text{const};$$
 (25)

 I_0 is proportional to the intensity of the wave at entry into the crystal and is conserved during propagation in it. Using the energy integral (25), we can reduce the number equations to three. Substituting (24) into (23), we obtain

$$\frac{d\Delta i}{d\xi} = q' i^{-} \sin 2\varphi, \quad \frac{di^{+}}{d\xi} = q' i^{-} \cos 2\varphi,$$

$$\frac{di^{-}}{d\xi} = -q' (\Delta i \sin 2\varphi + i^{+} \cos 2\varphi).$$
(26)

The phase difference of the wave A_x and A_y will be useful. It can be easily determined from the values of i^+ and i^- . Writing A in the form of complex quantities with phases $\Psi_x(\xi)$ and $\Psi_y(\xi)$,

$$A_x = |A_x| \exp[i\psi_x(\xi)], A_y = |A_y| \exp[i\psi_y(\xi)],$$

we have

$$i^{+} = 2 \frac{|A_x A_y|}{I_0} \cos(\psi_x - \psi_y),$$
$$i^{-} = \frac{2|A_x A_y|}{I_0} \sin(\psi_x - \psi_y)$$

and accordingly for the phase difference we have



$$\Delta \psi = \psi_{x} - \psi_{y} = \arctan(i^{-}/i^{+}).$$

Equation (26) was solved numerically with the initial conditions

$$\Delta i \Big|_{z=0} = -1, \ i^+ \Big|_{z=0} = 0, \ i^- \Big|_{z=0} = 0,$$

which are equivalent to the incidence of an extraordinary wave on the sample (Fig. 1).

From the numerical computation we obtained $i_x(\xi) = A_x A_x^*/I_0$ for the propagation of a wave inside the sample (Fig. 3). For the computation we took the parameters $\mu' = 3$ and 10, q = 10, which corresponds to a MBBA nematic liquid crystal with thickness L ~ 15 μ m and w ~ 26 and 300 cm/sec/cm. Figure 4 shows the phase difference $\Delta \psi(\xi)$ to within π for the same parameters. Figures and 4 indicate that $\mu' >> 1$ for saturation, i.e., when $\varphi = \pi/2$ in most of the volume, the intensity and phase difference inside on the free-surface side also tends to a constant, linear dependence. Otherwise ($\mu' \sim 1$), the wave characteristics are rather involved functions of the spatial variable ξ .

Figures 5 and 6 show the values of Δi and $1 + i^+$ at exit from the sample as functions of μ' for various values of q'. The values of Δi was chosen because it is usually more convenient to measure the intensity difference in an experiment.

The quantity $1 + i^+$ is important because it corresponds to the interference pattern that can be observed if the polarization plane is rotated in one of the components of the wave and the beams are superposed. Thus,

$$(|A_{r}|e^{i\varphi_{r}} + |A_{v}|e^{i\varphi_{v}})(|A_{r}|e^{-i\varphi_{v}} + |A_{v}|e^{-i\varphi_{v}})/I_{0} = 1 + i^{+}.$$

This quantity is of interest to us in regard to convenience of measurements. As is seen from Figs. 5 and 6, the desired relations have a rather complicated behavior. The greatest trouble is that they are nonmonotonic. During processing of the experimental data this circumstance can cause indeterminacy in the determination of μ' and q'from the measured values of Δi and $1 + i^+$.

The above computations thus establish a relation between the mechanical changes caused in the shear flow of a NLC layer by the motion of the upper free surface as a result of an aerodynamic flow and the changes in the diagnostic light beam transilluminating the liquid-crystal medium.

This treatment enables us to image a scheme for specific measurements. One result of the work is as follows. Using a diagnostic beam and getting two independent characteristics of the transmitted wave at the exit, we can measure, e.g., the velocity $v_y(L)$ of the upper boundary of the NLC layer, which cannot be determined without knowing the thickness L of the coating, which is why the second parameter of the transmitted wave was necessary.

Technically, this means that the emerging beam is presplit into two parts, One part passes through a plarizer, with axis oriented in the x direction, and the other part also passes through a polarizer, but with its axis oriented in the y direction. One beam is taken from each pair and the intensity difference Δi is measured. The beam remaining after the polarization plane is rotated in one of them come together and give us the interference intensity $1 + i^+$.

In summary, the result of our calculations reduce to the solution of a direct problem, establishing a relation between the mechanical parameters of the shear flow of a NLC layer with the optical response of a system, subjected to the action of an aerodynamic flow.

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