# Low-frequency local rotation of the molecules of nematic liquid crystals under the action of an acoustic shear wave and of an electric field

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The phenomenon of local rotation of the molecules of liquid crystals under the action of an acoustic shear wave and of an electric field is discovered, described, and investigated experimentally. The result is a continuous generation of disclinations with Frank index $m = -1$, moving in the plane of the specimen coaxially with respect to each other. In the plane of the specimen there is formed a system of such elements, distributed over the sites of a square lattice with a large period ~0.3 cm.

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In the study of the action of shear waves on oriented nematic liquid crystals (NLC), various authors have noted a manifestation of effects of the first order, expressed in a change of the director orientation at a frequency equal to twice the frequency of the shear waves,[1] and also effects of the second order, which produce a stationary orientation of the director.[2] Since for shear oscillations of the substrate the first-order effects attenuate rapidly with increase of frequency, $\delta n \sim \omega^{-2}$, the second-order effects become important at high frequencies and large amplitudes. All the principal observed acoustic-optical properties are a consequence of them.[3] Appropriate calculations of the director orientation and estimates of the contributions of these effects to the acoustic-optical effect have been made by Nagai and Iizuka[4] and by Dion and Jacob.[5]

The purpose of the present paper is to investigate secondary phenomena of director orientation during action of an acoustic shear wave on a LC, and in the presence of additional effects of director reorientation by an electric field, for example by virtue of the Freedericksz effect under the condition of rigid coupling of the molecules with the substrate.

## METHOD AND PRINCIPAL RESULTS OF THE EXPERIMENTS

A system of the sandwich type formed the basis of the experiments described below. The liquid-crystal layer was located between an acoustic radiator and a fused-quartz plate, on which was deposited a conducting transparent layer of $SnC_2$. The thickness of the layer was fixed by mylar spacers. The second electrode was sputtered on to the acoustic radiator. The source of acoustic shear waves used was a crystal of bismuth germanate, $Bi_{12}GeO_{20}$, of [100] cut. This choice was determined by two facts that are important for experiments with NLC. The first is the equality of the velocities of ultrasound in NLC and in a $B_{12}GeC_{20}$ crystal; the second, the possibility of obtaining significant amplitudes of the oscillations because of the large piezoelectric moduli of bismuth germanate, as high as $6 \cdot 10^{-6}$ cgs esu. The measurement-cell construction under consideration provided for simultaneous action of an acoustic shear wave even in the presence of molecule-reorientation effects in an electric field. This possibility was determined by the equality of the capacitive reactances $r_C$ and resistances $r_0$ of the NLC and of bismuth germanate: for spacer thickness $5 \sim 2 \cdot 10^{-3}$ cm for MBBA, and for acoustic-radiator thickness 0.5 cm, $r_C(\mathrm{NLC}) \sim r_C(Bi_{12}GeO_{20})$, and also $r_0(\mathrm{NLC}) \sim r_0(Bi_{12}GeO_{20})$.

In the experiments described above, nematic liquid crystals of the following compounds were used: MBBA, MBBA+ EBBA mixture, EBBA, and LC 440 mixture. To prescribe a planar orientation of the director, the substrates were polished in a single direction with M1 powder and diamond paste. The $Bi_{12}GeO_{20}$ crystal was polished along crystallographic direction [001] or [110]; this made it possible to determine the effect of the director orientation on the character of the interaction of acoustic waves and the NLC. The change of director orientation under the action of an electric field and of an acoustic shear wave was studied by an optical-polarization method, with a polarizing microscope provided with a spectral attachment that contained photometric apparatus. The time-variable light fluxes were followed, recorded, and processed with a memory oscillograph S8-2. The sources of the alternating and dc voltages $U_k$ and $U_x$, were a G3-33 and B5-50 generator, respectively. The sample was placed in a thermostated chamber. The temperature was recorded with a temperature regulator RT-3; the accuracy of fixing of the temperature was 0.1°. A copper resistance, $R = 47\ \Omega$ at $T = 20\,^\circ$C, served as temperature detector. The resonance frequency of the piezoelectric transducer was determined by the equivalent-resistance method.

All the results obtained can be summarized as follows. On application of an alternating voltage to a homeotropically oriented layer of nematic MBBA, a reorientation of the layer occurs at voltage ~10 V as a result of attainment of the Freedericksz threshold. With increase of the voltage on the cell being studied, and with attainment of the value $U \sim 40$ V, a system of disclinations, with Frank index $m = -1$, appears in the sample [Fig. 1(a)]. This structure is nonstationary in time, and at local points of the $xy$ plane the director ro-

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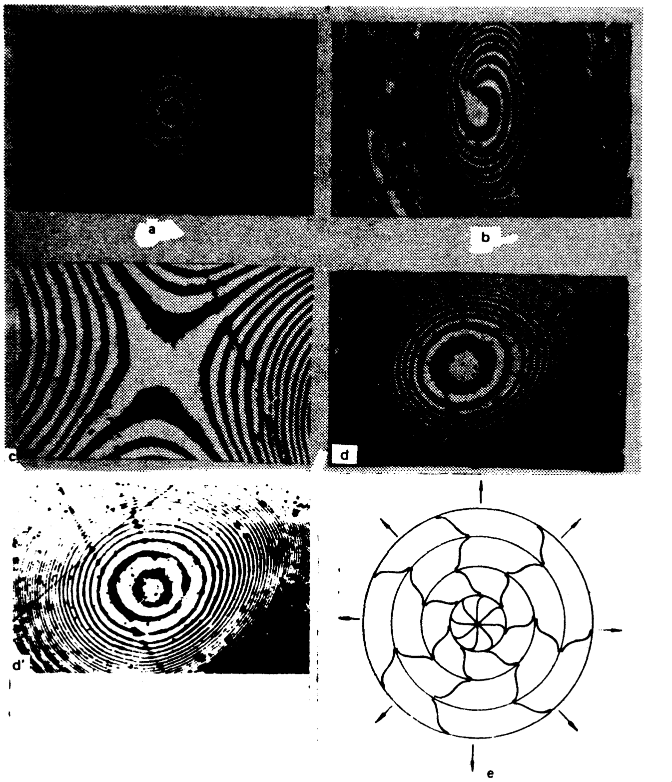


FIG. 1. Optical picture of local rotation of molecules, observed in a polarizing microscope (nicols crossed), magnification 10 ×. a—$U_k$ = 60 V, $U_x$ = 20 V; b—"magic spiral" of Meyer at $U_k$ = 60 V, $U_x$ = 20 V; c—junction of four elements of the vortex lattice; d and d', pictures of disclinations at time interval $\Delta t \sim 1$ s; e—orientation of molecules and of disclinations.

tates with a certain frequency $\Omega$ that depends linearly on the applied voltage (Fig. 2). The rotation process is not accompanied by displacement of the center of mass of the molecules.

Figure 3 shows diagrams that illustrate the local rotation of the molecules in the sample. The most convincing experiment demonstrating the presence of rotation of the molecules is the following. For a fixed voltage, it is possible to select a frequency of rotation of the polarizer such that the picture of the disclinations will be motionless. This frequency is equal in absolute value to the frequency of rotation of the director. A general picture of the distribution of the director rotation over the plane of the sample is shown in Fig. 3. It

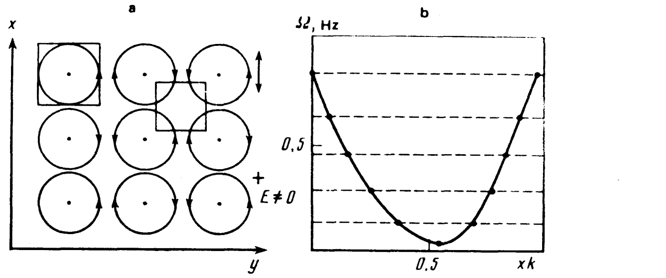


FIG. 3. Distribution of vortices (a) and of frequency of rotation of molecules (b) in the plane of the crystal. The squares show the correspondence with Figs. 1(a) and (b).

is typical that the distribution of angular velocities of rotation of the director in the plane of the sample has the form of a square lattice. But the angular velocity varies along the $x$ axis according to the law $\Omega \sim \cos^2 kx$ and is independent of the coordinate $y$ [Fig. 3(b)].

As a rule, when local rotation of the director occurs, disclinations with Frank index $m = -1$ are formed; but formation of disclinations with Frank index $m = +1$ is possible. In this case, the rotating picture is reminiscent of the Meyer magic spiral treated in the monograph of de Gennes[6] [Fig. 1(b)].

Generally speaking, the picture of concentric rings, moving radially, represents a set of coaxial disclinations with Frank index $m = -1$, generated after each period of rotation of the director. "Runoff" of disclinations and annihilation of them occur by two processes: by annihilation of disclinations at a point of junction of four elements of the lattice [Fig. 1(c)], and by annihilation of disclinations with opposite signs $m = \pm 1$ at great distances from the center of an individual vortex. These processes are illustrated in Figs. 4(a) and (b). The importance of the second process consists in the fact that it apparently determines the saturation of the velocity of rotation of the director by the applied voltage (Fig. 2), since the rate of annihilation of disclinations is independent of the applied voltage.[7]

Local rotation of the axes of the molecules occurs also in nuclei of NLC that form during the NLC-isotropic liquid phase transition. Here also the velocity of rotation depends linearly on the voltage, but the optical picture of the rotation, observed in a polarizing micro-

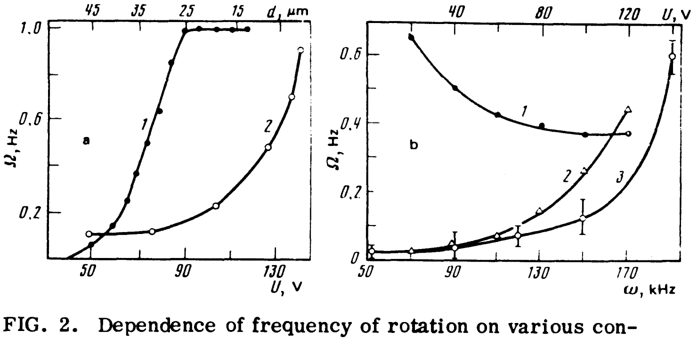


FIG. 2. Dependence of frequency of rotation on various conditions. a—voltage $U = U_k + U_x$ (1) and thickness $d$ (2). b—voltage $U_x$ when $U_k$ = const (1), $U_k$ when $U_x$ = const (2), and frequency $\omega$ of the ultrasonic wave (3) when $v_0^2 \cos^{-2}\psi$ = const.

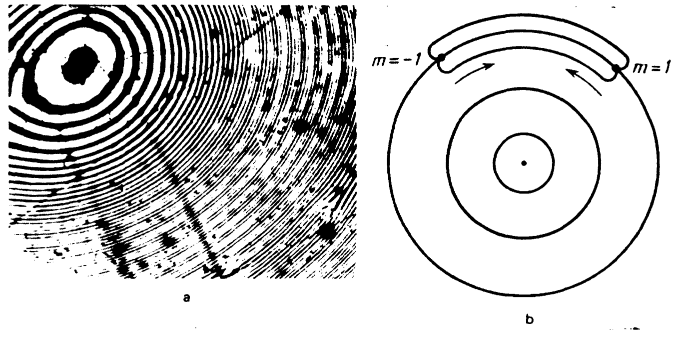


FIG. 4. Disclinations with Frank indices $m = \pm 1$ in the process of annihilation.

scope, is reminiscent of Lehmann's diagrams for cholesteric liquid crystals (CLC).[8] But in the case considered, in contrast to CLC, a temperature gradient does not lead to significant changes of the velocity of local rotation of the molecules.

The following necessary condition for manifestation of the phenomenon of local rotation of the director consists in the vanishing of the moment of elastic forces on the surface. A proof of this, for example, is the absence of the effect for planar orientation of the director at the surface. In this case, what occurs is the typical picture of a spatially periodic deformation or "rolls" structure. Such structures have been studied earlier, for example by Davis and Chambers[9]; therefore there is no point in dwelling on their description. The effect is also absent in NLC with positive dielectric anisotropy and homeotropic orientation of the director. But it was possible to observe spatially periodic structures at large intensities of the shear waves.

Since the above-considered phenomenon of local rotation of molecules in NLC with negative dielectric anisotropy is independent of the initial orientation of the director with respect to the wave vector of the shear wave, which is determined by the components $n_x$ and $n_y$ of the director, in NLC with positive dielectric anisotropy it is possible to observe local rotation in sections with dimensions of the order of the thickness of the crystal under study. This phenomenon occurs as a result of the appearance, upon application of a constant electric field parallel to a homeotropically oriented director, of domains with a cylindrically symmetric distribution within them of the components $n_x$ and $n_y$ of the director. The action of an acoustic shear wave on such a layer of NLC causes a phenomenon of local rotation of the director similar to that described above. A vortex lattice is formed in the crystal, with generation of disclinations with Frank index $m = +1$ (Fig. 5), and with dimensions of the individual elements of the order of the thickness of the crystal.

Further change of the velocity of the shear wave leads to a breakdown of the stability of the vortex lattice and to formation of oscillatory structures, representing a system of parallel two-dimensional disclinations, which cannot be stable[6] and which collapse with formation of the original vortex lattice (see Fig. 5). The period of oscillation of such structures varies linearly with the angular velocity of the director (see Fig. 2). Apparently the nature of the instability of a vortex lattice in this case is similar to the instability of a vortex lattice of helium II or to Kármán streets. The corresponding estimates of instability are given in the monograph of Putterman[10] and the paper of Tkachenko.[11]

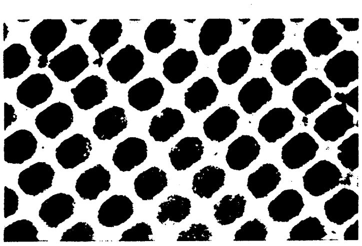

FIG. 5. Vortex lattice in NLC with $\varepsilon_a > 0$: magnification 250×; $U_x = 5$ V, $U_k = 20$ V.

## DISCUSSION OF EXPERIMENTAL RESULTS

Thus a very essential condition for observation of the phenomenon of local rotation of the director is orientation of the molecules at the boundary. With initial homeotropic orientation of the molecules and with development of the $B$ effect, i.e. with reorientation of the molecules in fields above the threshold, $U > U_c$, and with strong boundary conditions, the surface moments vanish. This condition is formulated as follows (the components $n_j$ of the director have the form $n_x = \cos\varphi\cos\psi$, $n_y = \sin\varphi\cos\psi$, $n_z = \sin\psi$, where $\varphi$ is the azimuthal angle and $\psi$ is the angle between the direction of orientation of the molecules and the $z$ axis)[6]:

$$\tau_{il} = e_{ijk} n_j \pi_{lk} = 0, \tag{1}$$

where $\pi_{lk} = \partial F/\partial g_{lk}$; $F$ is the free energy of the NLC, determined below; $g_{lk} = \partial n_l/\partial x_k$; and $e_{ijk}$ is the Levi-Cività symbol. From (1) we have in expanded form

$$n_x \pi_{zy} - n_y \pi_{zx} = 0 \tag{1'}$$

or

$$\left.\frac{\partial\varphi}{\partial z}\right|_{z=\pm a} = 0. \tag{1''}$$

If the depth of penetration of the elastic wave into the NLC exceeds the sample thickness, the problem of the motion of the director is two-dimensional, and this considerably simplifies the interpretation of the observed phenomena. We shall consider the complete system of equations that describe the motion of the director and of the centers of inertia of the NLC molecules. It consists of three equations[6]: the Navier-Stokes equation

$$\rho\frac{\partial v_i}{\partial t} + \rho v_k \frac{\partial v_i}{\partial x_k} = -\frac{\partial P}{\partial x_i} + \frac{\partial \sigma_{ik}'}{\partial x_k}; \tag{2}$$

the equation of motion of the director

$$[\mathbf{n}\times\mathbf{h}] = \mathbf{\Gamma}, \tag{3}$$

where $\mathbf{h} = \delta F/\delta\mathbf{n}$ is the molecular field,

$$F = -\frac{1}{2}\int\left\{K_{11}(\operatorname{div}\mathbf{n})^2 + K_{22}(\mathbf{n}\operatorname{rot}\mathbf{n})^2 + K_{33}[\mathbf{n}\times\operatorname{rot}\mathbf{n}]^2 + \frac{\varepsilon_a}{8\pi}(\mathbf{E}\mathbf{n})^2\right\}dV,$$

$K \sim K_{11} \sim K_{22} \sim K_{33}$ are the moduli of elasticity, and $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$ is the dielectric anisotropy of the NLC; and the equation of continuity

$$\frac{\partial\rho}{\partial t} - \operatorname{div}(\rho\mathbf{v}) = 0. \tag{4}$$

Here $\rho$ is the density, $P$ is the pressure, $x_i$ are the components of the vector displacement, $\sigma_{ik}'$ is the viscous stress tensor, and $v$ is the velocity of the centers of inertia. The viscous stress tensor $\sigma_{ik}'$ is represented as follows:

$$\sigma_{ik}' = \alpha_1 n_j n_p A_{jp} n_i n_k + \alpha_2 n_i N_k + \alpha_3 n_k N_i + \alpha_4 A_{ik} + \alpha_5 n_i n_j A_{jk} + \alpha_6 n_k n_j A_{ji},$$

$$A_{ik} = \frac{1}{2}\left(\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i}\right), \quad N = \frac{d\mathbf{n}}{dt} - \frac{1}{2}[\operatorname{rot}\mathbf{v}, \mathbf{n}], \tag{5}$$

where $\alpha_1, \ldots, \alpha_6$ are the Leslie coefficients of viscosity. Furthermore, the moment of the frictional forces due to the velocity of rotation of the director is

$$\Gamma=[\mathbf{n}\times(\gamma_1\mathbf{N}+\gamma_2\hat{A}\mathbf{n})],\quad \gamma_1=\alpha_3-\alpha_2,\quad \gamma_2=\alpha_3+\alpha_2. \tag{6}$$

Since we are interested in second-order processes, the equations of motion simplify and take the form (when $q \gg k$, where $q$ is the wave vector of the vortex lattice shown in Fig. 4 and where $k$ is the wave vector of the shear wave)

$$\rho v_k\frac{\partial v_i}{\partial x_k}\approx\frac{\partial\sigma_{ik}'}{\partial x_k},\quad \Gamma=[\mathbf{n}\times\mathbf{h}],\quad \operatorname{div}\mathbf{v}=0. \tag{7}$$

With allowance for the geometry of the problem, the following relations are valid in the interior of the crystal, $z\sim\delta_{eff}\sim(\eta/\rho\omega)^{1/2}$:

$$\rho v_x\frac{\partial v_x}{\partial x}\approx\gamma_2\left(\frac{\partial^2 n_x^2}{\partial x\,\partial t}+\frac{\partial^2(n_x n_y)}{\partial y\,\partial t}\right), \tag{8a}$$

$$\frac{\partial^2 n_y^2}{\partial y\,\partial t}+\frac{\partial^2(n_x n_y)}{\partial x\,\partial t}\approx 0, \tag{8b}$$

$$K\frac{\partial^2\psi}{\partial z^2}+\frac{\varepsilon_a}{2\pi}E^2\psi\approx 0,\quad \sin\psi\sim\psi. \tag{8c}$$

Hence when $v=v_0\cos kx\cos\omega t$, we get from (8a) and (8b)

$$\frac{\partial^2 n_x^2}{\partial x^2}-\frac{\partial^2 n_y}{\partial y^2}\approx Ct,\quad C=\rho v_0^2 k\gamma_2^{-1}\sin 2kx. \tag{9}$$

Here we have omitted a weakly oscillating term $C\cos 2\omega t$. Then for $q\gg k$ the solution of Eq. (8) has the form

$$n_x=(Ctr)^{1/2},\quad n_y=(1-Ctr)^{1/2},\quad r=[(x-na/2)^2+(y-ma/2)^2]^{1/2}; \tag{10}$$

here $na/2$ and $ma/2$ are the coordinates of the centers of the vortexes. Then the angular velocity of rotation of the director is

$$\Omega_{max}\approx 2\rho\gamma_2^{-1}kqv_0^2\sin 2kx\cos^{-2}\psi. \tag{11}$$

Since $\cos^2\psi=1-\sin^2\psi\sim U/U_c$, $\Omega\sim kU^2$, which describes the experiment qualitatively sufficiently well. Furthermore, on taking into account the estimates

$$\gamma_2\sim 0.8\ P,\quad v_0\sim\omega x_0\sim d_{133}\omega Ud^{-1}\sim 0.1U,\quad \rho\sim 12\ \mathrm{cm}^{-3}.$$

we get the sufficiently good quantitative agreement $\Omega\sim 1.6\cdot 10^3 U^2$ (see Fig. 2); here $d_{133}$ and $d$ are, respectively, the piezoelectric modulus and the thickness of the $Bi_{12}GeO_{20}$ crystal.

Besides the solution (10) considered above, the system of equations (2)–(4) has a second independent solution, resulting from Eq. (3). It describes, in general, disclinations unstable in time. In the case of two-dimensional disclinations, such instability manifests itself in annihilation of disclinations with opposite indices. The corresponding equation of state has the form

$$\frac{\partial^2\varphi}{\partial x^2}+\frac{\partial^2\varphi}{\partial y^2}-\frac{1}{D}\frac{\partial\varphi}{\partial t}=n\pi\delta(r-ut),\qquad D=K_{11}/\gamma_1,\quad r=(x^2+y^2)^{1/2}, \tag{12}$$

where $\delta(r-ut)$ is the Dirac delta function and where $u$ is the velocity of motion of the cores of the disclinations.

The solution of this equation is treated in detail in the paper of Imura and Okano.[12] In agreement with the experimental results set forth above, it describes the appearance and annihilation of disclinations with Frank indices $m=\pm 1$ at great distances from the center of an individual element of the vortex lattice, as is illustrated in Figs. 4(a) and (b). The generation of disclinations is a consequence of the relation (11). In fact, the instantaneous picture of identical phases of rotation of the director, $\varphi_0=\Omega t_0$, depends on the distance from the center of each element of the lattice. For a complete rotation $\varphi_0=l_\pi$, $l=0,1,2,\ldots$ and $\nu=0$, the relation between the dimensions of the rings is $r_l\sim l^{1/2}$. This corresponds to the relation that results from Fig. 1. Generation of disclinations with Frank indices $m=\pm 1$, shown in Figs. 4(a) and (b), corresponds to lag of the director rotation in neighboring regions $l$, $l+1$ by phase $\varphi_0=\pi$.

Thus a shear wave, during simultaneous action of an electric field on a homeotropically oriented liquid crystal with negative dielectric anisotropy, $\varepsilon_a<0$, causes low-frequency local rotation of the director. The observed effect finds explanation within the framework of the theory of nonlinear interaction of a shear wave and of the director under strong boundary conditions that keep the surface moments equal to zero.


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