Instability of planar texture of a cholesteric liquid crystal in an electric field

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Experiments carried out on nemato-cholesteric mixtures demonstrated that the threshold voltage U_{th} and period λ of instabilities of the "conducting regime" in a planar texture of a cholesteric liquid crystal with a pitch P_0 of the order of the cell thickness L were oscillatory functions of L. This was attributed to oscillations of the induced helix pitch P about its equilibrium value as a function of L, i.e., to the existence of Grandjean bands appearing due to orientation of the liquid crystal by the cell walls.

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The stability of a cholesteric liquid crystal (CLQ) layer in an electric field applied parallel to the cholesteric helix axis has been investigated theoretically[1–3] and experimentally.[4–6] It has been shown that for either sign of the dielectric anisotropy $\Delta\varepsilon$, spatially periodic deformations appear in a planar structure of a CLQ at a threshold voltage U_{th}. If $\Delta\varepsilon < 0$, these deformations are due to the electrical conductivity in a CLQ,[2–4] whereas if $\Delta\varepsilon > 0$, we can have both dielectric and conducting mechanisms.[3,5,6]

By analogy with a nematic liquid crystal (NLQ), the appearance of instabilities in the $\Delta\varepsilon < 0$ case is due to the anisotropy of the electrical conductivity and is a threshold process. The low-frequency threshold voltage U_{th} and the period of the resultant deformation λ are related to the parameters of the liquid crystal by[3]

$$U_{th}^2 = \frac{8\pi^3}{\varepsilon_\perp}\frac{\sigma_\parallel+\sigma_\perp}{\sigma_\parallel-\sigma_\perp}\left(\frac{3}{2}K_{22}K_{33}\right)^{1/2}\frac{L}{P_0}, \quad (1)$$

$$\lambda^2 = \left(\frac{3}{2}\frac{K_{33}}{K_{22}}\right)^{1/2}P_0L, \quad (2)$$

where $\sigma_\parallel$ and $\sigma_\perp$ are the components of the electrical conductivity parallel and perpendicular to the director; K_{22} and K_{33} are the Frank elastic moduli for torsion and longitudinal bending, respectively; P_0 is the equilibrium value of the helix pitch; L is the thickness of the cell enclosing the liquid crystal. Equations (1) and (2) are derived on the assumption that $L \gg P_0$ and they ignore the difference between the real (induced) helix pitch P and the equilibrium value P_0, which results from the orienting influence of the cell walls.

Experiments have confirmed that the theoretically predicted dependences $U_{th} \propto \sqrt{L/P_0}$ and $\lambda \propto \sqrt{LP_0}$ on condition that $L \gg P_0$, and then the threshold deformation of a CLQ is in the form of a quadratic network.[4,6]

Extrapolation of Eq. (1) to high values of P_0 (dilute nemato-cholesteric mixtures) or to low values of L, where the inequality $L \gg P_0$ is no longer satisfied, gives values of U_{th} much smaller than the threshold voltages of the domain instability in an NLQ.[7,8] In view of this it seemed interesting to investigate the threshold characteristics of the instability in a planar texture of a CLQ with a pitch P_0 of the order of the cell thickness L. We investigated primarily CLQ's with $\Delta\varepsilon < 0$, because the electrooptic properties of these substances (with $P_0 \sim L$) represented the so far uninvestigated intermediate case between the usual dynamic scattering of light and the dynamic "memory" scattering. The instability in CLQ's in the case of L equal to $\frac{1}{2}P_0$, P_0, and $\frac{3}{2}P_0$ had been observed earlier,[5] but detailed investigations of the threshold characteristics have not yet been made.

EXPERIMENTAL METHOD

Our liquid crystals were a mixture of a nematic substance with a cholesteryl caprinate (up to 5% by weight). Our NLQ was a mixture of azoxy compounds (mixture A[1)]) in the temperature range from -10 to $+74$ °C. The starting materials had resistivities in the range 10^{12}–10^{13} Ω · cm and were used without further purification. Controlled electrical conductivity and its anisotropy $\sigma_\parallel/\sigma_\perp$ were assured by doping mixture A with tetrabutylammonium bromide (0.01% by weight), which gave $\sigma_\parallel/\sigma_\perp = 1.8$ and $\sigma_\perp = 3\times10^{-9}$ Ω^{-1} · cm^{-1} at room temperature. This doping did not alter the components of the permittivity: $\varepsilon_\parallel = 4.9$ and $\varepsilon_\perp = 5.3$. The addition to mixture A of small amounts of cholesteryl caprinate did not affect the electrical conductivity and permittivity components, which was checked by special measurements. We assumed that the elastic properties and viscosity were also unaffected.[7,8]

It was established that the equilibrium helix pitch P_0 of CLQ samples prepared in this way containing a proportion C by weight of cholesteryl caprinate in mixture A was described by the relationship $P_0C = 0.14 \pm 0.03$ μ. Measurements of the threshold voltage and deformation period were carried out in planar and wedge-shaped cells. Glass walls with deposited transparent electrodes were rubbed by the Chatelain method. The thickness of the gap in the cells was set by Teflon spacers and could be varied from 6 to 200 μ. The homogeneity of the air gap over the whole area of a planar cell was checked in monochromatic light using equal-thickness fringes and it could be established to within 0.5–2 fringes, which corresponded to an inhomogenity of the cell thickness in the range 0.1–0.5 μ. The actual thickness was deduced from the interference spectrum of the cell transmission obtained with a recording spectrophotometer.[9]

A wedge-shaped gap between two glass plates was

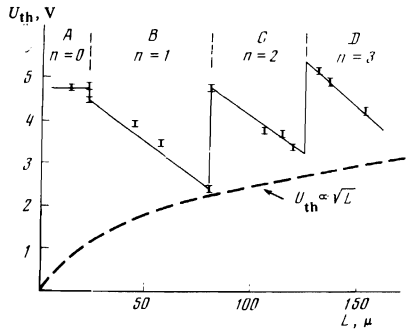

FIG. 1. Dependence of the threshold voltage U_{th} on the thickness L of a planar cell. Mixture A + 0.1% cholesteryl caprinate ($P_0 = 115\ \mu$). The continuous curve is based on the experimental results and the dashed curve on the Helfrich–Hurault theory.

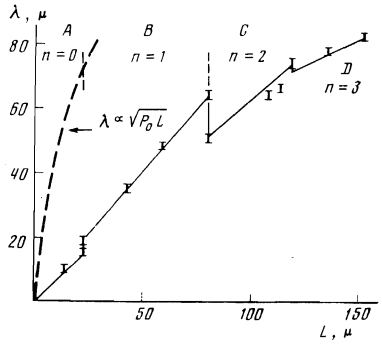

FIG. 3. Dependence of the deformation period λ on the thickness of a planar cell. Mixture A + 0.1% cholesteryl caprinate ($P_0 = 115\ \mu$). The continuous curve is based on the experimental results and the dashed curve on the Helfrich–Hurault theory.

formed by the use of Teflon spacers of different thickness in such a way that the directions of rubbing b of the glasses (parallel to one another) were perpendicular to the thickness gradient in the wedge. The wedge angle α was deduced from the distance between equal-thickness fringes in monochromatic light of a mercury lamp ($\lambda = 0.546\ \mu$): $\alpha = \lambda/2l_1$. The helix pitch P_0 was determined from the distance l_2 between Cano–Grandjean disclinations, corresponding to a sudden change by unity in the number of half-turns in the helix which can be fitted in the local thickness of the wedge (first-order disclinations): $P_0 = 2\alpha l_2$.

The instability threshold voltage U_{th} and the deformation period λ were determined at 20 Hz (which was much less than the critical frequency for the conducting regime[3,4]) using a polarizing microscope (the polarizer was parallel to b). At this frequency the injection effects were unimportant. The value of U_{th} was determined to within 0.1 V.

RESULTS AND DISCUSSION

A typical dependence of U_{th} on the thickness L of a planar cell is shown in Fig. 1. We can clearly see the regions within which U_{th} decreases approximately linearly with increasing thickness, separated by sudden switching of U_{th}. Similar regions are observed also in the dependence of U_{th} on the local thickness L of a wedge-shaped cell (Fig. 2). These regions are separated by Cano–Grandjean disclinations and they can be seen also in Fig. 3, which gives the dependence of the deformation period λ on the thickness of a planar cell L.

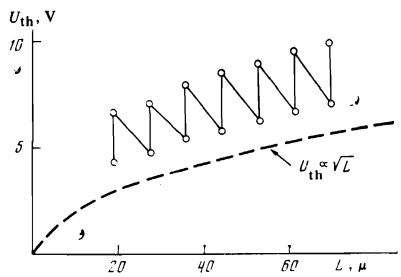

FIG. 2. Dependence of the threshold voltage U_{th} on the local thickness L of a wedge-shaped cell. Mixture A +1% cholesteryl caprinate ($P_0 = 16\ \mu$). The continuous curve is based on the experimental results and the dashed curve on the Helfrich–Hurault theory.

The nature of instabilities is different in each region (Fig. 4). In region A the deformation is manifested by bands perpendicular to the rubbing direction b; they are practically indistinguishable from Williams domains observed in the initial (without the addition of cholesteryl caprinate) nematic mixture. In region B the bands are parallel to b. In region C they are perpendicular to b. Both perpendicular and parallel band orientations are observed in region D (the orientation depends on the depth of focusing of the microscope). In the succeeding regions the deformation forms a square network.

The existence of these regions can be explained by a model describing the appearance of an induced helix pitch P (different from the free pitch P_0) in cells with two orienting planar surfaces. The dependence of the induced pitch P on the distance between the orienting surfaces (in wedge-shaped or planar cells) is shown in Fig. 5. The dependences are calculated on the assumption that the directions of rubbing are parallel and that in any cell thickness L we can fit an integral number of half-turns of the helix, i.e., $P = 2L/n$, where $n = 1, 2, 3, \ldots$ is the number of half-turns in the helix. It is also assumed that the number of half-turns changes abruptly by unity half-way between two thicknesses corresponding to the equilibrium pitch. Region A corresponds to a stressed NLQ in which there is no helical structure. When the number of half-turns increases, the deviation of the induced pitch P from the equilibrium value P_0 decreases, so that if $L \gg P_0$, the pitch in the cell can be regarded as having the equilibrium value.

The nature of the deformation shown in Fig. 4 confirms the proposed model. In region A (untwisted CLQ) the type of deformation is the same as in the original nematic substance (Williams domains). In region B (corresponding to one half-turn of the helix) the bands are parallel to the rubbing direction b because at midpoint of the cell thickness the director is perpendicular to b,[5] and so on. Region A represents a stressed NLQ or an untwisted CLQ. Within this region the value of U_{th} is independent of L and P_0 (only the length of region A depends on P_0) and it is identical with U_{th} of the original NLQ (4.7 V).

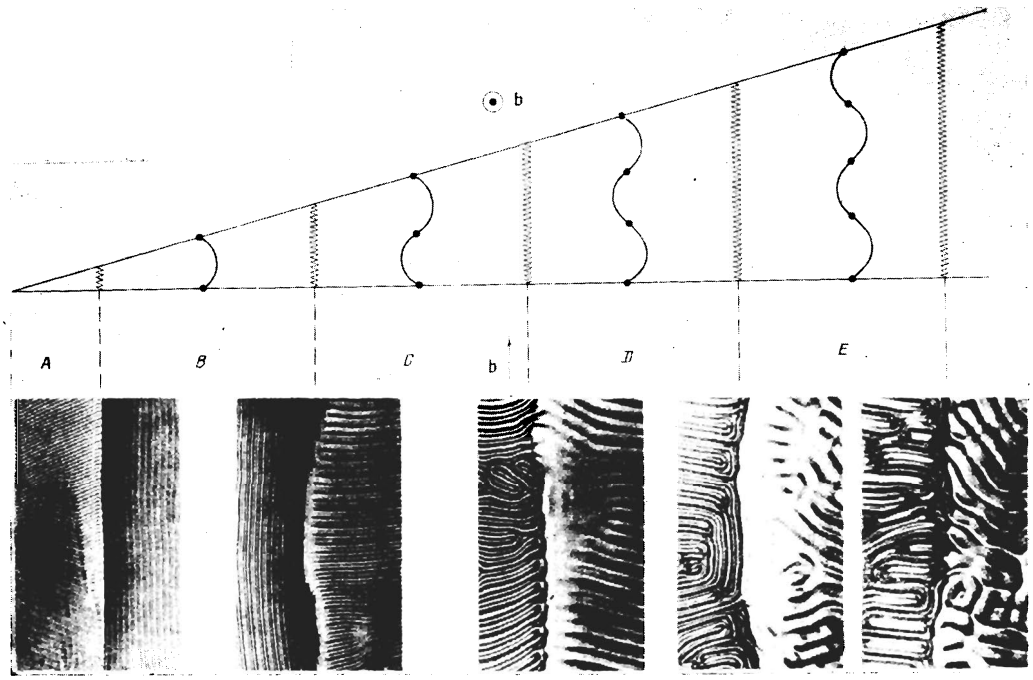

FIG. 4. Threshold deformation patterns near Cano–Grandjean disclinations. Mixture A + 0.1% cholesteryl caprinate (P_0 = 115 μ) in a wedge-shaped cell. The photographs were recorded using the same magnification and voltages across the cell at which deformation was excited on both sides of a disclination.

The jump in U_{th} between regions A and B is of opposite sign compared with jumps between other regions and its magnitude is less. It should be noted that the jump of 2.2–2.5 V between other regions, beginning from B, is independent of L (Figs. 1 and 2), which is surprising because the deforming tension in the helix is weak when $L \gg P_0$. The existence of this jump is responsible for the frequently reported nonreproducibility of the results of measurements of U_{th} for $P_0 \ll L$ in planar cells when the slightest change in the cell thickness L causes a transition from one region to another accompanied by a jump in U_{th}. The existence of irremovable disclinations in some planar cells is also due to the fact that there are several regions with different values of n in the same cell (see L = 22.6 μ and L = 80 μ in Fig. 1).

In region B the value of U_{th} reaches its minimum (U_{th}^{min} = 2.5 V) near the boundary with region C and the value of U_{th}^{min} increases slightly with decreasing value of P_0. Nevertheless, for a given cell thickness L the addition of a CLQ to an NLQ in such a concentration that the pitch P_0 of the resultant mixture satisfies the relationship $P_0 \gtrsim 4/3L$, reduces considerably U_{th} of an NLQ.

It is interesting to note that the turbulent motion visible under a microscope appears in region A (and also in the original NLQ) at voltages exceeding U_{th} by 20–30%, whereas in other regions turbulence is observed for $U = (1.5\text{–}2)\,U_{th}$.

The present authors are not aware of any theory describing the appearance of instabilities in a CLQ with a pitch of the order of the cell thickness L allowing for the existence of an induced pitch P differing from the equilibrium value P_0. We shall assume that the physical cause of the reduction in U_{th} with increasing L within one region is the tendency for a deformed helix to return to its equilibrium state. Thus, a stretched helix tends to become detached from the walls and, therefore, the required destabilizing moment of the electric field decreases. The opposite is true of a compressed helix.

An exact calculation of U_{th} clearly requires that the expression for the free energy[3] should contain an additional term describing the deformation of the helix by the cell walls. Nevertheless, our results can be explained qualitatively on the basis of the Helfrich–Hurault theory[1–3] assuming that in Eqs. (1) and (2) we can replace the equilibrium pitch with the real (induced) pitch $P = 2L/n$, and the elastic moduli K_{22} and K_{33} can be replaced with the effective (apparent) moduli K_{22}^* and K_{33}^*. Then, Eq. (2) can be rewritten in the form

$$\lambda = \left(\frac{3}{2}\frac{K_{33}^*}{K_{22}^*}\right)^{1/2}\sqrt{\frac{2}{n}}\,L. \qquad (3)$$

It is clear from Eq. (3) that when the number of half-turns is increased, the value of λ should decrease discontinuously and the magnitude of a jump should decrease with increasing n. Within one region (n = const) the value of λ should rise linearly with L and the coefficient of proportionality should decrease with increasing n, i.e., the ratio K_{33}^*/K_{22}^* should be independent of L. When the above substitutions are made, Eq. (1) becomes

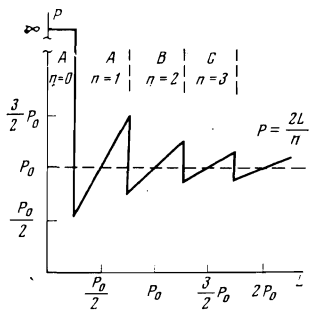

FIG. 5. Dependence of the induced helix pitch P on the cell thickness L.

$$U_{th}^2 = \frac{8\pi^3}{\varepsilon_\perp}\frac{\sigma_\parallel+\sigma_\perp}{\sigma-\sigma_\perp}\left(\frac{3}{2}K_{22}^*K_{33}^*\right)^{1/2}\frac{n}{2}. \quad (4)$$

This relationship describes the jumps of U_{th} resulting from variation of n (with the exception of the $B \to A$ transition corresponding to complete untwisting of a CLQ into an NLQ). The dependence of U_{th} on L within one region can be explained simply by assuming that K_{22}^* and K_{33}^* vary with L. Thus, the oscillatory nature of the dependences of U_{th} and λ on L can be explained qualitatively.

Experimental studies of the Fréedericksz transition and untwisting of cholesteric helices is an electric field gave the values of $K_{33} = 1.0\times10^{-6}$ dyn and $K_{22} = 0.3\times10^{-6}$ dyn. Figures 1–3 include the dependences of U_{th} and λ on L calculated on the basis of Eqs. (1) and (2) for the equilibrium value of the pitch. We can see that the Helfrich–Hurault theory (derived for $L \gg P_0$) describes only qualitatively the average dependence of U_{th} and λ on L for $L \sim P_0$.

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[1)]Mixture A consists of two parts of p-n-butyl-p'-methoxyazoxybenzene and one part of p-n-butyl-p'-heptanoyloxyazoxybenzene.

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Nonlinear Raman interaction between first and second sounds in liquid helium II

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The nonlinear Raman interaction between first and second sounds in liquid helium II, i.e., the parametric excitation of second sound by first sound in a resonator, is considered. An expression is obtained for the threshold first-sound intensity. The intensities of the stationary waves are found. The deviation from exact synchronism of the interacting waves and the difference between the frequencies of these waves and the natural frequencies of the resonator are taken into account. The stability of the obtained solutions is investigated.

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It is well known that liquid helium II begins to manifest nonlinear properties when sufficiently intense first- and second-sound waves propagate in it. Thus, Osborne[1] observed the formation of second-sound shock waves, the theory of which is given in Khalatnikov's paper,[2] where first-sound shock waves are also considered. The formation of shock waves is, in essence, a self-action of waves, and is not connected with nonlinear mixing of waves of different nature. Another type of nonlinear interaction—the so-called nonlinear Raman interaction connected with the nonlinear intermixing of first- and second-sound waves—is also possible. In the present paper we consider one of the examples of the latter interaction, namely, the parametric excitation of second-sound waves at the expense of the exciting first-sound waves. This process is, in particular, of interest from the point of view of the generation of high-frequency second sound. Such an interaction was considered in[3] for the case of traveling waves. However, since the threshold first-sound intensity starting from which second-sound wave excitation becomes possible essentially depends on the attenuation of the waves,[3] a more favorable case for the experimental observation of the process in question is the case of waves in a resonator, since losses in a resonator with a sufficiently high Q are less than in traveling waves. In view of this, in the present paper we consider the parametric excitation of second sound by first sound for the case of waves in a resonator.

In solving the problem, we proceed from the nonlinear equations of the hydrodynamics of a superfluid liquid,[4] transformed into a form suitable for our problem.[3] These equations are coupled, nonlinear wave equations for pressure and temperature. Since we can assume the nonlinearity of the medium to be sufficiently