Disclination mechanism of instability in nematic-cholesteric mixtures

R. I. Mints, E. V. Kononenko, and E. G. Aksel'rod

S. M. Kirov Ural Polytechnical Institute
(Submitted 7 July 1980)
Zh. Eksp. Teor. Fiz. **80**, 1122–1134 (March 1981)

An optical polarization method is used to study the instability of the diffuse flow of a cholesteric liquid crystal in a plane capillary containing a nematic, under homeotropic boundary conditions. It is established that upon attainment by the flow of a critical flow velocity, when the pitch of the helix is approximately equal to the thickness of the capillary, a transition occurs from one low-energy configuration to another, by distortion of the director field on the lines of flow. The experimental data are corroborated by numerical calculations carried out within the framework of the Leslie-Ericksen approach. A model of the structure of the lines of flow is considered. A mechanism is proposed for the origin, development, and relaxation of instability of plane flow of a nematic-cholesteric mixture; the mechanism is based on the formation, interaction, and transformation of different types of axial disclinations of oppsite sign.

PACS numbers: 47.20. + m, 47.60. + i

Study of the influence of linear defects of structure, of the disclination type, on the dissipative properties of liquid crystals (LC) is promising for description of the hydrodynamics and structure of LC in a state of turbulence and of dynamic scattering.[1] But the role of disclinations in the processes involved in the transition to turbulence is unclear.[2] Turbulence of the flow of LC may be due to cooperative generation of defects or to a continuous distribution of defects of small strength.[3]

The purpose of the present paper is to study disclination effects in the mechanism of the origin, development, and relaxation of instability of plane flow of LC with a helical structure. The optical analogy between the disclination structures that are formed during formation of a nematic-cholesteric mixture (NCM) and the textures that originate during field effects suggest that distortions of quasinematic layers, due to the appearance and development of a disclination structure in the mixture, are similar to the instabilities that accompany the process of field untwisting of the cholesteric spiral.[4]

For the "temporal unfolding" of the changes of distribution of the director field during a transition to turbulence, we have used the method of the concentration gradient.[5] We have considered the morphological and topological parameters of linear defects identified in NCM during diffusion of a cholesteric LC into a plane capillary containing a homeotropically oriented nematic. The presence in such a mixture of a substantial concentration gradient of the optically active component guarantees diffusive flow with varying conditions of twist. The action of boundary forces, perpendicular to the plane of flow, leads to the appearance of disclination lines and of singular points, which interact vigorously with each other and exert an influence on the flow properties.

1. METHOD AND EXPERIMENTAL RESULTS

An optical polarization method was used to investigate the diffusion of cholesteryl peargonate (CP) into a plane cell filled with nematic *p*-methoxybenzylidene butylaniline (MBBA). The measurements were made at a controlled temperature of 22 °C. A standard sandwich cell was used, whose thickness ($L = 10$ μm) was prescribed by teflon spacers and was measured by an interferometric method[6] with a scanning measurement microscope ORIM-1, with accuracy 0.1 μm. The spacers were cut out in such a way that a narrow (0.5 mm) streak of CP could diffuse linearly from the edge of the slide into the MBBA. The initial homeotropic orientation of the MBBA in the plane of the capillary was achieved by treatment of the glass in accordance with Berreman.[7]

The pitch P of the helix in the NCM at small CP contents ($C < 1$ wt. %) was determined by the Grandjean-Cana method with wedge-shaped cells. For $1\% < C < 5\%$, the pitch was estimated from the relation $P = \lambda_{max}/\langle n \rangle$, where λ_{max} is the light wavelength corresponding to the maximum of the selective reflection by the plane texture; this wavelength was measured by a microspectrophotometric method,[8] by use of an attachment to the polarizing microscope, which consisted of a monochromator, a photoelectronic multiplier, and a recording system operating by count of single-electron pulses; $\langle n \rangle$ is the average index of refraction, determined on an Abbe refractometer. Use of the polarization accessories provided with the MBI-9 microscope, with a precision focusing micromechanism (0.001 mm), made it possible to identify structural changes occurring in different layers of the thin (10 μm) capillaries.

The initial stages of diffusion in the NCM (after the beginning of the capillary sucking in of the CP) are accompanied by the appearance of specific periodic structures, constituting series of optically inhomogeneous bands, which are arranged along the direction of diffusion and are separated by regions with moving concentration-determined Grandjean disclinations[9] (Fig. 1a, left part). The presence of Grandjean disclinations moving in the direction of the concentration gradient of the CP, which is characteristic of the plane texture of the NCM, and also the direct observation of motion of the material in the system of bands, suggest that

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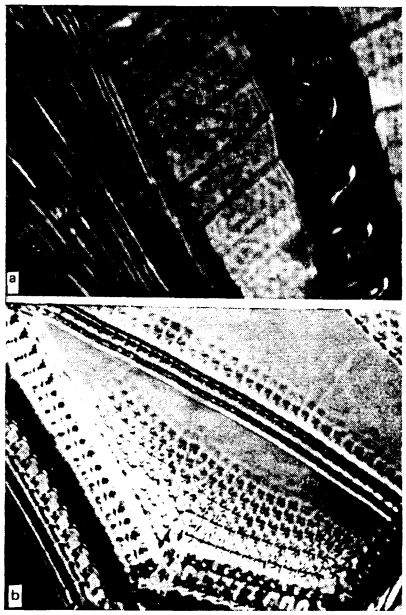

FIG. 1. Instability of plane flow of NCM with homeotropic boundary conditions (×400): a—wave-shaped distortions in the system of flow lines; b—transition to two-dimensional distortion of quasinematic layers.

in the middle of the layer there is plane flow due to diffusion. The structure of the lines of flow is found to be unstable. After 9 ± 1 min, at a distance 0.8 ± 0.1 mm from the beginning of the diffusion zone, wave-shaped distortions originate in the system of bands, developing over several seconds (Fig. 1a, right part). The distortions begin at the boundary of the first and second Gradjean zones and gradually capture the flow lines over the whole diffusion zone. Both the distortions and the deformed flow lines have a number of peculiarities: each of them is morphologically connected with a definite second-order Grandjean disclination; to Grandjean disclinations with small numbers correspond lines located in the center, with large numbers along the periphery, of the systems of bands. The contrast of the lines of flow is practically unchanged on rotation of the polarizer, whereas the optical path difference, for unchanged position of the polarizer, changes considerably near the lines of flow and diminishes abruptly (approximately by a factor two) at the line itself.

The successive morphological changes in the middle plane reduce to a transition from a one-dimensional distortion of quasinematic layers to a two-dimensional spatially periodic distortion: after 10–12 min, in the region of periodic distortions there is observed a spiral twisting of the cholesteric planes, and the Grandjean disclinations relax to a system of mutually orthogonal focal lines, superposition of which forms a spatially modulated structure of square-grid type (Fig. 1b). A similar structure forms also near the regions of helical twisting of the lines of flow; here the appearance of the grid always occurs to one side of the line.

Further diffusion of the CP and lowering of its concentration gradient in the MBBA are accompanied by a transition to a stable domain structure in the region of twisting, and to a system of polygonal fields on the focal lines. The topological and morphological features of disclinations and other defects in these structures have been considered in a previous paper.[4]

We shall estimate the parameters of the diffusion flow (the pitch of the helix and the velocity of flow) in the range where the wave-shaped distortions of the banded structure originate. For this purpose, using the method of Ref. 10, we shall describe the diffusion of CP into a plane cell containing MBBA. The coefficient D of diffusion of a cholesteric mesophase into a nematic, at normal temperature, does not exceed 10^{-6} cm^2/sec.[11] If t is the instant corresponding to the appearance of distortion in the system of bands ($t \sim 9$ min), then the length of the diffusion path $[\sim(Dt)^{1/2}]$ is found to be much smaller than the extent of the cell in the direction of diffusion (17 mm); therefore the influence of the transverse boundaries can be neglected. For simplicity, we shall consider one-dimensional diffusion from a linear source of constant strength (a thin streak of CP on the edge of the slide) into a semi-infinite medium (MBBA) with zero initial concentration of the diffusing component.

For a constant and concentration-independent diffusion coefficient, the solution of the equation of Fick's second law for the given initial and boundary conditions will have the form

$$C(x,t)=C_0[1-\mathrm{erf}\,(x/2(Dt)^{1/2})], \tag{1}$$

where $C_0=\mathrm{const}$ is the initial concentration of CP in MBBA for $x=0$ (pure CP); D is the average diffusion coefficient over time t.

By using the experimentally established relation between the pitch P of the spiral in the NCM and its CP content,

$$PC=\gamma; \quad \gamma=0.13 \ \mu\mathrm{m\ at} \quad C\leqslant 0.05 \ \mathrm{wt.\%}, \tag{2}$$

and taking account of the relation

$$L=NP/2, \tag{3}$$

where N is the number of half-turns of the cholesteric helix spanned by the local thickness ($N=1,2,3,\ldots$) one can determine D, C, and P_0.

On the microphotographs obtained at a fixed instant of time, we measured the distance from the beginning of the diffusion zone to the first concentration-determined Grandjean disclinations ($N=1,2,3$). Thereby, values of x and t were determined by a known cell thickness (10 μm) for a discrete set of spiral pitches (3) and corresponding concentrations (2). On substituting C, x, and t in (1) and using tables of values of the error integral, we found a mean value $D=8.9\cdot10^{-7}$ cm^2/sec, which practically coincides with that obtained under similar conditions.[11] Then for the region of origination of wave-shaped distortions in the system of bands (at $x\approx0.9$ mm, $t\approx9$ min), the concentration of CP in the NCM according to (1) is $C\approx1\%$; that is, the helix pitch $P\approx13$ μm.

The velocity of the diffusion flow in this region was

determined by means of an eyepiece micrometer, from the velocity of motion of optical microinhomogeneities (see the points A, A' etc. in Fig. 4) present in the system of flow lines. The mean value of the flow velocity was found to be 1.2 $\mu m^2/sec$.[1)]

The character and the kinetics of the textural changes described above indicate that plane flow of the NCM, during diffusion into a nematic with homeotropic boundary conditions (HBC), is unstable at a certain stage. In the static case, the homeotropic texture of a nematic LC during introduction of a cholesteric component remains stable up to a certain critical twist.[12] The reasons for and the mechanism for bringing about the instability, under conditions of diffusion flow, are not completely clear within the framework of the usual model, which considers deviations of the axis of the cholesteric helix in a direction parallel to the walls of the cell, with formation of a texture of the "fingerprint" type (FPT).[13]

The banded structure of the flow lines in Fig. 1a is morphologically similar to the FPT, but under homeotropic conditions the FPT is stable; furthermore, the observed distortions cannot be explained by periodic variations of the index of refraction in the plane of flow, caused by regular variations of the angle of inclination of the molecules to the helicoidal axis. Actually the lines, first, are due to disclinations in the plane of flow (Fig. 1a), and the distances between the lines, in contrast to the FPT, are not related to the value of the local pitch of the NCM (whether forced or equilibrium); second, the lines are optically inhomogeneous, i.e., they consist of alternating light and dark sections, separated by singular points; third, the optical-anisotropy peculiarities mentioned above, on the lines, are also not characteristics of the FPT.[14]

Thus the wave-shaped instability of the diffusion flow cannot be interpreted as the result of a periodic deformation of quasinematic planes of the FPT under the action of a secondary flow.[15] In the present paper, a possible mechanism is proposed for the origin, development, and relaxation of the instability; it is based on the interaction of dynamic paraxial disclinations of opposite sign.

2. ENERGY OF TWIST AND CONDITIONS FOR INSTABILITY

It is natural to suppose that the reason for the instability of plane flow is a transition of the molecules from one low-energy configuration to another, induced by the change of twist conditions. In Sec. 1 it was shown that to the stage of diffusion under consideration corresponds a "thin specimen" in which $L/P \sim 1$. It is known[16] that for thin specimens, the energy of distortion of a cholesteric helix is determined chiefly by the energy of twist.

Following Press and Arrott,[17] in order to calculate the energy of twist under conditions of stationary plane flow and to determine the corresponding low-energy configuration (LEC) of the molecules, we introduce a coordinate system XYZ such that the Z axis is perpendicular to the walls of the cell ($-L/2 \leqslant z \leqslant L/2$) and the X axis coincides with the direction of flow. In consequence of the anisotropy of the viscosity, deviations from the X direction may occur in the flow; but for small velocities of flow (of the order of a few $\mu m^2/sec$), when the effective viscosity is independent of the velocity,[18] these deviations may be neglected.

In the one-constant approximation ($K_{11}=K_{22}=K_{33}=K$), the energy of twist will be

$$E=\frac{1}{2}K\int dz\left[\left(\frac{\partial n_x}{\partial x}\right)^2+\left(\frac{\partial n_y}{\partial y}\right)^2+\left(\frac{\partial n_z}{\partial z}\right)^2+2q'n_x\frac{\partial n_y}{\partial z}-n_y\frac{\partial n_z}{\partial z}\right)+q^2\Big]. \tag{4}$$

Here n_x, n_y, n_z are the components of the director, of constant length ($n^2 \equiv 1$), and q is the wave vector of the spiral, $q = 2\pi/P$.

When a diffusion flow is present in the NCM, there acts on the director field a twist, which is determined by the usual expression for the molecular field,

$$\mathbf{h}=\gamma_1\left(\frac{d\mathbf{n}}{dt}-[\omega\times\mathbf{n}]\right)+\gamma_2\mathbf{n}A_{xy}, \tag{5}$$

where ω is the angular-velocity field, $\omega = \frac{1}{2}\mathrm{rot}\mathbf{V}$; $\mathbf{V}$ defines the flow-velocity field; the viscous parameters γ_1 and γ_2 are linear combinations of the Leslie coefficients, describing the orientational effects of the flow: $\gamma_1=\alpha_3-\alpha_2, \gamma_2=\alpha_3+\alpha_2$; A_{xy} is the symmetric part of the velocity-gradient tensor,

$$A_{xy}=\frac{1}{2}\left(\frac{\partial V_y}{\partial x}+\frac{\partial V_x}{\partial y}\right). \tag{A}$$

At constant flow velocity, in a stationary state, the material derivative $d\mathbf{n}/dt$ of the local director is zero. Then the expression for the molecular field of the flow, oriented along the X axis, will have the form

$$\mathbf{h}=\frac{dV}{dz}(\alpha_3\mathbf{n}_x+\alpha_2\mathbf{n}_z). \tag{6}$$

Minimization of the twist energy (4) with respect to the possible deviations of $\mathbf{n}$, with allowance for the molecular field (6), gives a system of equilibrium equations under conditions of constant diffusion current,

$$\begin{aligned}\frac{\partial^2 n_x}{\partial z^2}&=\lambda n_x+2q\frac{\partial n_y}{\partial z}-\frac{\alpha_2}{K}\frac{dV}{dz}n_z,\\ \frac{\partial^2 n_y}{\partial z^2}&=\lambda n_y-2q\frac{\partial n_x}{\partial z},\\ \frac{\partial^2 n_z}{\partial z^2}&=\lambda n_z-\frac{\alpha_3}{K}\frac{dV}{dz}n_y,\end{aligned} \tag{7}$$

where λ is a Lagrangian multiplier.

We choose a parabolic law of variation of the velocity of diffusion flow with cell thickness, as the most probable for simple flow between two parallel plates with HBC[17]:

$$V(z)=V_{max}[1-(2z/L)^2], \tag{8}$$

where V_{max} is the velocity of the midplane in the cell. Accordingly, the linear velocity gradient

$$\left|\frac{dV}{dz}\right|=V_{max}\frac{8z}{L^2} \tag{9}$$

will be greatest at the boundary surfaces and zero at the middle layer of the NCM.

With use of (3), integration of the equilibrium equations (7), under the conditions (9), enables us to cal-

culate the variation of the twist energy (4) with the pitch of the NCM for a fixed diffusion velocity and a constant number of half-turns N. We shall give numerical estimates. We take $K=10^{-11}$ N,[19] $\alpha_2/K=3.2\cdot10^6$, $\alpha_3/K=6.4\cdot10^4$,[20] $L=10$ μm = const, $z=1$ μm (near the middle plane). We specify $N=1,2,3$ and $V=0$–25 μm^2/sec in steps of 2 μm^2/sec. Solving (7) by the method of finite differences,[21] we get, in coordinates EPV, three families of curves for the variation of the twist energy in the first to third Grandjean zones. The results of the calculation for $N=1$ and for $N=2$ are given in Fig. 2a. The cases $N=1$ and $N=2$ correspond to the experimental situation of instability opposite the boundaries of the first and second Grandjean zones. In Fig. 2a one can distinguish a definite region of pitches and flow velocities, to one side of which the family of curves for $N=2$ is located above the family of curves for $N=1$, and therefore the lower-energy configuration is that with $N=1$, whereas on the other side the lower-energy configuration is that with $N=2$.

Thus in a dynamic range of helix pitches and flow velocities corresponding to the spatial line of intersection of the families of energy curves $E(P,V)$ with $N=1$ and with $N=2$, the configuration of molecules for $N=1$ is an equilibrium configuration but not a stable one, since there is a configuration $N=2$ of lower energy under the given conditions. This allows us to regard the indicated interval ($P=10$–13 μm, $V=0$–22.5 μm^2/sec) as a region of instability of plane flow in NCM, within which there can occur a transition from one low-energy configuration to another. The family of energy curves with $N=3$ (not shown in Fig. 2a) is located above the curves $N=1$ and $N=2$ for all values of P and V; the curves with $N=4$ go even higher, etc. This is a reflection of the fact that the energy of distortion of the quasinematic planes increases with increase of the degree of twist of the NCM.[22]

Pictures of the director distribution in a layer of NCM, for $N=1,2,3$, are shown schematically in Fig. 2b. The regions of distortion ($\pm\xi$) indicate homeotropic arrangement of the boundary molecules under the action of boundary forces, since the velocity and the orienting

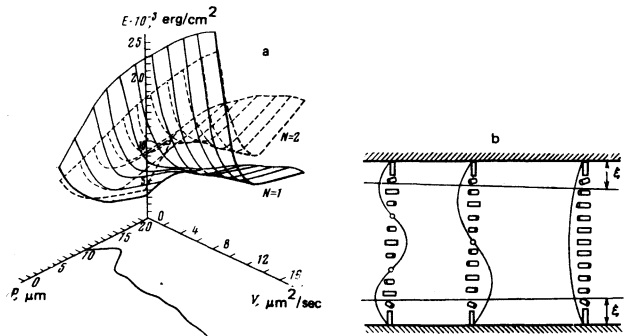

FIG. 2. Determination of the parameters of instability of plane flow. a—change of twist energy on transition from the first to the second Grandjean zone, at $P=13$ μm and $V=2$ μm^2/sec. b—schematic sketch of the distribution of molecules in the instability region. The curve in the PV plane is the projection of the spatial line of intersection of the energy surfaces for $N=1$ and for $N=2$.

influence of the flow according to (8) are smallest at the boundary surfaces. Over a thickness $L-2\xi$ the texture of the NCM may be regarded as plane (ξ decreases rapidly with increase of the ratio L/P).

It follows from Fig. 2 that the instability of plane flow is accompanied by a transition of the molecules in the middle regions from an orientation predominantly along the direction of flow ($N=1$) to an orientation predominantly perpendicular to the flow ($N=2$), since the helix pitch of the NCM decreases constantly in the diffusion process.

When $V>22.5$ μm^2/sec, for all values of P, the energy curves with $N=1$ are located below the curves with $N=2$; that is, at high velocities of flow (of the order of several tens of μm^2/sec) the orienting action of the flow is effective, so that the molecules in the central layers are always arranged predominantly parallel to the flow. Such a situation occurs in the capillary sucking in of CP. In the case of shear-induced flow of the NCM into a wedge-shaped cell with HBC, at flow velocities 10–15 μm^2/sec, lines corresponding to an odd number of half-turns of the cholesteric helix are stable.[17] With decrease of V to a few μm^2/sec, there occurs a rotation of the lines perpendicularly to the flow. A critical velocity V_{cr} is determined by the ratio of the thickness of the wedge and the pitch of the spiral and is attained when $L/P\approx1$. As in our case, the twist of the molecular structure in the instability region is determined by the expression

$$\Gamma=[\mathbf{hn}]=\frac{dV}{dz}[(\alpha_2 n_z^2-\alpha_3 n_x^2)+\alpha_3 n_x n_y-\alpha_3 n_y n_z]. \qquad (10)$$

In MBBA with small chiral additions, $\alpha_2\gg\alpha_3$,[23] therefore the terms in (10) that describe twist about the Y and Z axes are of order n_z and nearly vanish in the central region. Reorientation of the director in these regions occurs primarily in the plane of twist.

The calculated values of P and V agree satisfactorily with the experimental parameters obtained in the preceding section. Thus the region of occurrence of wave-shaped distortions of the banded structure is actually a region of instability of diffusion flow of the NCM and is due to a tendency of the LC molecules to orient themselves predominantly perpendicular to the flow within a certain range of flow velocities and of spiral pitches.

3. STRUCTURE OF THE LINES OF FLOW

The numerical estimates presented are approximate, since the calculation of the low-energy configuration was based on a number of assumptions: the flow velocity and the elastic moduli do not remain constant during the change of composition of the NCM, and the relation $n^2=1$ is not satisfied in view of the appreciable birefringence on the lines of flow. It must be supposed that the plane orientation of the molecules during their rotation in the region of instability is preserved mainly for the middle of the LC layer; in other planes, and especially on approach to the boundary surfaces under the influences of HBC, deviations from the horizontal configuration increase.

The onset of instability is accompanied by a high flow

velocity, which effectively orients the long axes of the molecules along the flow. Using the system of coordinates adopted in Sec. 2, we can write the components of n in the form

$$n_x=\cos\theta(x,z), \quad n_y=0, \quad n_z=\sin\theta(x,z), \tag{11}$$

where $\theta(x,z)$ is the angle between the z axis and the XY plane; here $\theta(x,z)=q(x,z)x$, where q is the wave vector of the spiral. Under HBC, $\theta(x,\pm L/2)=n\pi/2$, where n is an odd number.

On substituting (11) in the expression for the free energy of the NCM.

$$E=\tfrac{1}{2}K[(\operatorname{div}\mathbf{n})^2+(\mathbf{n}\operatorname{rot}\mathbf{n}+q_0)^2+[\mathbf{n}\times\operatorname{rot}\mathbf{n}]^2], \tag{12}$$

we find that minimization of the elastic energy is achieved for values of $\theta(x,z)$ that satisfy the two-dimensional Laplace equation

$$\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}\right)\theta=0. \tag{13}$$

The solution of (13) determines lines of axial disclinations distributed in a horizontal plane along the Y axis. The distance between the lines along the horizontal is determined by the relation

$$\theta_n=\frac{1}{2}\sum_n \psi_n, \tag{14}$$

where ψ_n is the angle between X axis and a straight line connecting the origin of coordinates with the disclination line.

Further refinement of the structure of the disclination lines can be carried out by use of the condition of the molecular field for satisfaction of the HBC. A calculation analogous to the calculation of Cladis and Kléman[14] shows that most stable configuration is one in which the molecules in the surface layer lie in a plane perpendicular to the glass and twist nonuniformly in the internal part of the specimen. Presence of HBC causes preferred dissociation of axial disclinations of the Grandjean type [also determined by an expression of the form (13)] into disclination pairs of the $\lambda\tau$ type.

Thus there is a basis for supposing that during formation of linear flow, when the velocity and the orienting action of the flow are large, the flow structure is determined by a series of disclination pairs of the $\lambda\tau$ type, consisting of continuous lines (λ) and singular points (τ). The structure of the flow lines is close to the structure of Grandjean walls, since it is likewise caused by a change of the conditions of twist in a layer.

4. DISCLINATION MODEL OF INSTABILITY

The experimental results obtained and the results of the calculations show that a model describing the observed effects must: 1) explain the peculiarities of the optical anisotropy of the structure (the small change of contrast on rotation of the polarizer, the abrupt decrease of the optical path difference between the lines of flow); 2) describe the reasons for and the mechanism of the distortion of cholesteric layers, which leads first to a wave-shaped distortion in the system of bands and then to a two-dimensional spiral twist; 3) take into account the fact that at all stages of distortional reconstruction, the lines of flow are morphologically connected with concentration-dependent Grandjean lines of the second order. The proposed disclination mechanism for the origination, development, and relaxation of the instability of plane flow of the NCM consists of the following.

1. The initial stages of the diffusion of CP into MBBA are characterized, on the one hand, by strong flow (of the shear type), which for sufficiently large flow velocities (of the order of tens of $\mu m^2/sec$) effectively orients n along the direction of flow; and, on the other hand, by the presence of considerable gradients of concentration and accordingly of the pitch of the spiral. Under conditions of plane flow, this leads to the formation of concentrational Grandjean disclinations. The data of Sec. 3, and also the connection of Grandjean disclinations of the second order, that is of disclination pairs $\lambda^+\lambda^-$,[24] with the flow lines allows us to suppose that the structure of the latter is also dependent on the appearance of a system of axial disclination lines of opposite signs.

A possible model of the structure of the lines of flow is represented in Fig. 3. The difference of the described structure from the usual Grandjean-Cano geometry consists in the presence, along with disclinations of the λ^+ and λ^- types, of τ^+ and τ^- disclinations, joined into pairs of the wall type, connected with disclination lines and point singularities. The appearance in the flow-line structure of axial τ^+ and τ^- disclinations turns out to be possible, because the line of a $\tau^+\tau^-$ pair is always parallel to the axis of the spiral of the cholesteric liquid crystal (perpendicular to the local director), and the molecules in the region of a τ line have a tendency to arrange themselves along it in order to decrease the core energy, whose value is high on the τ disclinations.

Furthermore, the structure of the λ and τ pairs is such that the distances between the lines of the defects and the corresponding singular points are related in a definite way to the pitch of the helicoidal structure ($\tau^-\tau^+-P/2$, $\tau^-\lambda^+-3P/4, \lambda^+\lambda^--P/2, \tau^+\lambda^--P/4$).[25] The presence of a considerable and rapidly changing gradient of the pitch in the direction of diffusion [from (1) and (2)].

$$\frac{dP}{dx}\sim\frac{P^2}{(\pi Dt)^{1/2}}e^{-x^2/4Dt}, \tag{15}$$

and also a certain anisotropy of the viscosity of the NCM in the initial stage of flow, when the flow velocity is re-

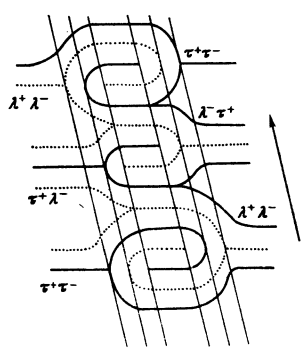

FIG. 3. Disclination model of the structure of lines of flow. The arrow shows the direction of the diffusion flow.

latively large, leads to the existence of a change of pitch within a system of lines. As a result, the structure of the flow lines is a set of pairs $3P/4, P/2, P/4$, located according to the change of gradient of the pitch.

The proposed model explains the morphological and optical-structural features, described in the preceding sections, of the band texture in the initial stages of diffuse flow. The existence of light and dark lines, the contrast of which is only slightly sensitive to rotation of the polarizer, is caused by depolarization and scattering of light by τ lines with singular cores. The abrupt change of the optical path difference is due, apparently, to the close agreement of the structure of the lines with the structure of Grandjean disclinations of the second order.[26 2)] The absence of correlation between the helix pitch and the period of the lines is a consequence of the close positioning of the cores of the τ lines in the structure.

2. With decrease of the flow velocity and of the helix pitch of the NCM, the structure of the lines of flow in the system of bands becomes unstable; and when the flow reaches a critical (P, V) region, there occurs a reorientation of the director to a direction in the flow plane perpendicular to the flow. The model being analyzed enables us to consider a mechanism of reorientation that describes the formation of wave-shaped distortions (see Fig. 1a). A schematic representation of the process is shown in Fig. 4.

A peculiarity of the disclination structure of a system of lines of flow is the fact that the pairs of disclinations that form it are complexes of zero strength,[27] and consequently they do not interact with each other in the configuration shown in Fig. 3. This means that for relatively large V and P, the system of bands is stable. It is known that the structures of disclinations of the λ and τ types are similar.[28] For λ and τ lines, there is a modification of the molecular arrangement such that it changes a τ disclination locally to the λ type. This transition is accomplished by rotation of the director through 90° near the line of the defect. Under conditions of diffuse flow, such a transition occurs especially easily, since the energy of the cores of τ disclinations is large, there are singular points on the lines of cores, and for a $\tau^+\tau^-$ pair there are even two discontinuities at a time in the direction of the molecules. The energy of a λ line is much lower, because the director field on the line of the defect experiences no singularity. Thus the rotation of the director to a direction perpendicular to the flow in the instability region is a consequence of the disclination reaction $\tau \rightarrow \lambda$.

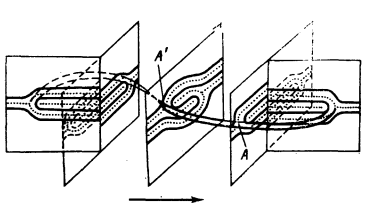

FIG. 4. Interaction of disclinations during the development of instability on lines of flow. For clarity, the figure has been turned 90° with respect to the preceding figure.

After the transformation $\tau \rightarrow \lambda$, the strengths of the lines acquire different signs; and since the state of distortion of the quasinematic planes to the sides of the line of the defect is already inhomogeneous. the complexes of zero strength relax. The further development of instability of plane flow is due to the association of pairs of disclinations that originate in the system of bands with concentration-dependent Grandjean disclinations of the second order, whose structure is described in the $\lambda^+\lambda^-$ model. From geometric consideration it is clear that lines of a $\lambda^+\lambda^-$ pair in Grandjean disclinations and λ lines in the instability region may exist in planes the distance between which does not exceed $P/8$. At small vertical distances between lines of the same strength, the tendency toward formation of pairs increases abruptly.[27] Therefore λ lines of opposite signs in the structure of the flow lines and Grandjean disclinations tend to associate into pairs in the instability region. This tendency leads to a turning of part of the λ lines along the Grandjean disclinations, so that interaction occurs with Grandjean lines of the second order; alternating packets of lines rotate, as is shown in Fig. 4.

Undoubtedly the distinction between two independent stages of development of the instability is to a certain degree arbitrary. Under actual conditions, the $\tau \rightarrow \lambda$ transition and the subsequent rotation of λ pairs occur practically simultaneously. Their successive accomplishment is possible, for example, as a result of helicoidal glide[29] of individual disclinations along the gradient of the flow velocity. A consequence of this process is the system of dynamic pairs of axial disclinations $\lambda^+\lambda^-$ depicted in Fig. 4. The partial depolarization and scattering of light by lines of pairs shows up optically as wave-shaped light and dark lines, whose periodicity correspond to the variation of the gradient of the NCM helix in the direction of diffusion.

3. Further decrease of V and P is accompanied by the specific effect of an increase of the influence of secondary flow, directed perpendicularly to the primary and caused by the spiral structure of the NCM. The peculiarity of the influence of secondary flow on the cholesteric planes in the region of the λ lines consists in the fact that the twisting of the planes on disclinations of the λ and τ types occurs in mutually perpendicular directions. This process is represented schematically in Fig. 5. If the twisting occurs on straight-line sections of λ pairs, double spiral lines appear (see Fig. 1b, left); deformation of quasinematic planes at bends and breaks of the λ lines shows up as a segmentation of the structure of the flow lines, with formation of an internal quasi-domain structure (see Fig. 1b, right). Formation of a system of focal lines and subsequent transition to a spatially periodic distortion at small L/P occur always to one side of the flow lines (see Fig. 1b). If we assume that in these stages of diffusion the structure of the lines of flow is determined by double loops of disclination pairs $\lambda^+\lambda^-$, then we can show that these loops bound regions with numbers of half-turns of the cholesteric helix that differ by one.[30] Then the threshold and period of the two-dimensional distortion, which, other things being equal,

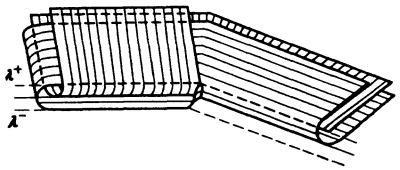

FIG. 5. Distortion of cholesteric layers on lines of disclinations during relaxation of instability.

are determined by the ratio L/P, will always be less to one side of the defect. This is confirmed by direct observations and measurements (see Fig. 1b).

Disclinations similar to Grandjean walls, but oriented perpendicular to the latter, are encountered in cholesteric LC with large pitch, placed in a wedge-shaped cell. Stieb[27] showed that the structure of such disclinations, as in the present work, is determined by complexes of disclination lines with zero strength. These complexes are formed by 90° rotation of the cores of λ lines joining into pairs; molecules in the region of nonsingular cores are arranged perpendicular to the local director. In our case, the inverse transition occurs.

In conclusion, we mention Ref. 31, in which it was discovered that the development of electrohydrodynamic instability of plane texture of NCM with negative dielectric anisotropy, in a field parallel to the axis of the helix, is due to origination and interaction of pairs of axial disclinations of the type $\lambda^+\lambda^-$ and occurs when $L \sim P$. The changes of the twist conditions here are analogous to those realized in our work. Probably the proposed disclination mechanism of instability has a quite general character in weakly twisted cholesteric systems.

[1)]The mean flow velocity was determined from the relation $V=Lx/t$ in order to make it possible to compare the results for different thicknesses of the capillaries and for fixed width of the gap. The first line in the concentration-determined Grandjean texture separates the regions of nematic phase and of mixture with cholesteric structure; therefore its velocity of motion corresponds to the velocity of the diffusion front.

[2)]On the Grandjean disclinations there is a substantial decrease of the path difference.

[1]P. de Gennes, J. Phys. (Paris) 35, L217 (1974).
[2]P. G. de Gennes, Physics of Liquid Crystals, Oxford Univ. Press, 1974, p. (Russian transl., "Mir", 1977, p. 228).
[3]J. Friedel, J. Phys. (Paris) 40, C3-45 (1979).
[4]R. I. Mints, E. V. Kononenko, and E. G. Aksel'rod, Kristallografiya 25, 989 (1980) [Sov. Phys. Crystallogr. 25, 567 (1980)].
[5]R. Mintz, E. Kononenko, and E. Akselrod, III Liquid Crystal Conf., Abstracts, Budapest 1979, p. D8.
[6]V. P. Gartshteĭn, N. A. Dudaev, and G. R. Ivanitskiĭ, in: Metody i tekhnika mashinnogo analiza biologicheskikh struktur (Methods and Technique of Machine Analysis of Biological Structures), M.: Nauka, 1972, p. 59.
[7]D. W. Berreman, Phys. Rev. Lett. 28, 1683 (1972).
[8]L. S. Agroskin and G. V. Papayan, Tsitofotometriya (Cytophotometry), L.: Nauka, 1977, p. 240.
[9]H. Kelker, Mol. Cryst. Liq. Cryst. 15, 347 (1972).
[10]H. Hakemi and M. M. Labes, J. Chem. Phys. 61, 4020 (1974).
[11]H. Hakami, and M. M. Labes, J. Chem. Phys. 63, 3708 (1975).
[12]F. Fischer, Z. Naturforsch. 31a, 41 (1976).
[13]L. M. Blinov and S. V. Belyaev, in: Kholestericheskie zhidkie kristally (Cholesteric Liquid Crystals), Novosibirsk, Siberian Branch Academy of Sciences, USSR, 1976, p. 69.
[14]P. Cladis and M. Kléman, Mol. Cryst. Liq. Cryst. 16, 1 (1972).
[15]U. D. Kini, J. Phys. (Paris) 40, C3-62 (1979).
[16]M. J. Press and A. S. Arrott, J. Phys. (Paris) 37, 387 (1976).
[17]M. J. Press and A. S. Arrott, J. Phys. (Paris) 39, 750 (1978).
[18]U. D. Kini, G. S. Ranganath, and S. Chandrasekhar, Pramana (India) 5, 101 (1975).
[19]W. Helfrich, Appl. Phys. Lett. 17, 531 (1970).
[20]J. Prost and H. Gasparoux, Mol. Cryst. Liq. Cryst. 22, 25 (1973).
[21]M. J. Press and A. S. Arrott, Mol. Cryst. Liq. Cryst. 37, 81 (1976).
[22]M. Kawachi, O. Kogure, S. Yoshi, and Y. Kato, Jpn. J. Appl. Phys. 14, 1063 (1975).
[23]A. Saupe, Ann. Rev. Phys. Chem. 24, 441 (1973).
[24]J. Rault, Phil. Mag. 28, 11 (1973).
[25]Y. Bouligand, J. Phys. (Paris) 33, 715 (1972).
[26]G. Malet and J. Martin, J. Phys. (Paris) 40, 355 (1979).
[27]A. E. Stieb, J. Phys. (Paris) 40, C3-94 (1979).
[28]J. Rault, J. Phys. (Paris) 33, 383 (1972).
[29]M. Kléman, in: Advances in Liquid Crystals, Vol. 1, ed. G. H. Brown, Academic Press, 1975, p. 267.
[30]Y. Bouligand, J. Phys. (Paris) 35, 959 (1974).
[31]T. Kohno, H. Miike, and Y. Ebina, J. Phys. Soc. Jpn. 44, 1678 (1978).

Translated by W. F. Brown, Jr.