

# Linear interaction of waves in liquid-crystal optics

V. V. Zheleznyakov, V. V. Kocharovskii, and V. V. Kocharovskii

*Institute of Applied Physics, USSR Academy of Sciences*

(Submitted 23 April 1980)

Zh. Eksp. Teor. Fiz. 79, 1735-1758 (November 1980)

The propagation of infrared and optical electromagnetic waves in smoothly nonuniform cholesteric liquid crystals is considered. Attention is called to the possibility of linear interaction of the waves in such a medium as a result of the inhomogeneity of the cholesteric helix (i.e., of the nonuniform rotation of the optical axes of the medium). In the first part of the paper (§§ 2-5) is introduced an interaction parameter that determines the wave-conversion effectiveness, the conditions are indicated for the onset of the conversion, and solutions are analyzed of two standard problems that describe the linear interaction in propagation of light wave in an inhomogeneous liquid-crystal medium. With these problems as examples, new possibilities of controlling the intensity and the polarization of light in liquid crystals are discussed. In the second part (§§ 6-10) is considered the connection between the critical field of a second-order phase transition in a liquid-crystal structure and the optical threshold of polarization cutoff of the light. These phenomena result from the reorientation of the director in an external electric or magnetic field. The optical thresholds for the planar twist structure, the Grandjean structure, and of the homotropic structure in a longitudinal field are theoretically determined, as well as for the cholesteric-nematic transition. The known experiments on polarization cutoff of light are explained.

PACS numbers: 78.70. — g, 61.30.Gd, 64.70.Ew

## § 1. INTRODUCTION

It is known<sup>1,2</sup> that in liquid crystals, despite partial or complete absence of spatial ordering, orientational ordering is preserved: the long axes of the molecules are aligned predominantly in a direction characterized by a unit vector  $L$ —the director. In liquid crystals of the cholesteric type, which contain molecules that have no mirror symmetry, the director  $L(z)$  describes a helical line with pitch  $g = 2\pi/h$  with a displacement along the  $z$  axis given by the wave vector  $h$  ("cholesteric helix"). The pitch of this helix can be controlled easily by placing the cholesteric crystal in an external electric and an external magnetic field or by varying the temperature, the pressure, and the chemical composition of the crystal.

From the point of view of electromagnetic wave propagation, such a crystal is an anisotropic medium with optical-axis orientation that varies in space. A homogeneous cholesteric with physical parameters that are constant in space (including the pitch  $g$  of the helix) is equivalent to a periodically inhomogeneous anisotropic medium. In such a medium, for a wave of frequency  $\omega$  propagating in the direction of the helix axis  $z$ , there exist four linearly independent solutions; they correspond to two normal waves traveling in the  $+z$  direction, and two normal waves in the opposite direction.<sup>3-6</sup>

In a smoothly inhomogeneous cholesteric whose physical parameters (helix pitch and other quantities that determine the refractive indices and the character of polarization of the normal waves) vary in space over a scale  $\Lambda \gg \lambda$  (where  $\lambda$  is the wavelength in the medium), the electromagnetic field equations have asymptotic solutions that determine the wave propagation in the geometrical-optics approximation. The field is represented as a superposition of independent geometrical-optics waves propagating along the helix axis. The polarization and the refractive indices of these waves are

uniquely determined by the local properties of the cholesteric [including the pitch of the helix  $g(z)$  at the given point] and do not depend on the concrete character of the inhomogeneity. We emphasize that the relation between the pitch  $g$  of the helix and the inhomogeneity scale  $\Lambda$  can be arbitrary. The indicated geometrical-optical waves in a smoothly inhomogeneous cholesteric, which are locally close to normal waves in a homogeneous cholesteric crystal, will be called helical waves (indicating thereby that they differ from the usually considered ordinary and extraordinary waves with refractive indices  $n_o$  and  $n_e$  in a homogeneous and anisotropic medium). The independent propagation of the helical waves means that the only optical effect is the geometrical-optical beats between the waves. This effect is indeed observed and can serve, e.g., to reveal phase transitions in liquid crystals by passing through the liquid-crystal layer light in the form of a superposition of normal waves.<sup>7,8</sup>

However, the geometrical-optics approximation may not hold if the refractive indices  $n_1$  and  $n_2$  of the helical waves propagating in the same direction become close in value. If the helical waves pass through a region in which the properties of the cholesteric vary significantly over the scale of the spatial beats between the waves

$$\Lambda \lesssim 2\pi/k_0 |\tilde{n}_1 - \tilde{n}_2| \quad (1.1)$$

( $k_0 = \omega/c$  and  $c$  is the speed of light in vacuum), then linear interaction between the waves can set in. This effect consists of a change in the ratio of the complex amplitudes of the helical waves passing through the indicated region. In particular, when a helical wave of one type is incident on the interaction region, two mutually coherent helical waves (with refractive indices  $\tilde{n}_1$  and  $\tilde{n}_2$ ) leave the region. It is clear that if a linear interaction is realized and leads to a mutual transformation of the helical waves, the polarization of the light passing through the liquid crystal can be substantially altered. The effectiveness of this linear transformation

depends on the inhomogeneity scale  $\Lambda$  and on the cholesterical-helix pitch  $g$  in the interaction region, so that variation of these quantities leads to a change in the optical polarization properties of the liquid-crystal layer.

The theory of linear interaction of light waves, which is developed in the present article, is of special interest for the determination of the ratio of the polarization cutoff of light and of the phase transition in liquid-crystal structures. Phase transitions between different conformations of nematics placed in a magnetic or electric field were first observed in 1927 by Fréedericsz and a group of co-workers<sup>9,10</sup> by optical methods. These transitions occur when the applied magnetic or electric field is changed and are due to the reorientation of the long axes of the liquid-crystal molecules as a result of the anisotropy of the diamagnetic and dielectric susceptibilities. The Fréedericsz transitions are therefore of second order,<sup>11</sup> and produce rather small changes in the optical properties of the liquid-crystal structure if the electric or magnetic field is only slightly stronger than the critical value. On the other hand, light cutoff calls for a substantial deformation of the structure; therefore the observed optical threshold, which is connected, e.g., with the cutoff of the polarized light, does not coincide with the critical point in the Fréedericsz transitions.

A similar situation, generally speaking, takes place also for phase transitions in cholesteric liquid crystals, and particularly for the cholesteric-nematic transition.<sup>12,13</sup> The investigation of the differences between the phase and optical thresholds was initiated relatively recently; the corresponding experiments for the nematic twist structure, which is most widely used, were performed only in 1971.<sup>14,15</sup> The differences indicated have not yet been investigated theoretically, notwithstanding the importance of this problem, which is raised in particular by the extensive use of measurements of threshold fields for the study of the physical properties of liquid crystals.

The linear-conversion phenomenon is extensively discussed in the theory of propagation of electromagnetic waves in a plasma (see, e.g., Refs. 16 and 17 and the bibliographies therein). At the same time, there is an obvious analogy between the two media, a plasma with a magnetic-field induction vector  $B_0$  that rotates in space, on the one hand, and a cholesteric liquid crystal, on the other. In both media we have rotation of the anisotropy axis, determined by the magnetic field  $B_0$  in the former medium and by the director  $L$  in the latter. This allows us to use, when considering linear interaction of light in liquid crystals, the research technique and the results obtained for the same effect in a plasma with a sheared magnetic field (see Ref. 5 and cf. Refs. 4 and 18). This analogy has prompted us to study linear conversion of waves in liquid crystals; to our knowledge such a phenomenon was heretofore not investigated theoretically as applied to liquid crystals. Of course, a plasma with a magnetic field differs substantially from a liquid crystal: the plasma with the magnetic field has optical activity (magnetic gyrotropy), whereas a liquid crystal can be locally regarded as a

uniaxial (with axis  $L$ ) crystal that has no optical activity. Therefore the onset of linear conversion of light in a smoothly inhomogeneous liquid crystal can be due only to rotation of the optical axes of the medium, i.e., it can take place only in crystals of the cholesteric type (see § 3). In a plasma, on the other hand, in addition to rotation of the magnetic field  $B_0$  in space, the reason for the wave interaction is the spatial inhomogeneity of its gyrotropic properties (cf. in this connection results of Refs. 5 and 17).

We consider below only uniaxial liquid crystals of the cholesteric type (in particular, twisted nematics), in which the magnetic and natural optical activities can be neglected. This choice allows us to simplify the exposition considerably; it is easy, however, to adapt the results to a smoothly inhomogeneous biaxial optically active medium with optical axes that rotate in space (cf. Ref. 5). The absorption in the crystals is not taken into account. Therefore the strong electron-absorption bands in the ultraviolet part of the spectrum call for a special analysis.<sup>19</sup>

## §2. HELICAL WAVES IN CRYSTALS OF CHOLESTERIC TYPE

We consider a uniaxial cholesteric characterized by a constant helix pitch  $g_0$  in the  $z$ -axis direction. To describe the normal waves  $\exp(i\omega t - ik \cdot r)$  propagating along the helix axis  $z$  in such a cholesteric, we introduce two coordinate systems, laboratory and local. The laboratory system  $x_0 y_0 z_0$  is Cartesian with a fixed orientation of the axes  $x_0$  and  $y_0$ ; the local system  $xyz$  is likewise right-handed and orthonormal, but its  $x$  and  $y$  axes follow the rotation of the cholesteric anisotropy axis  $L$  (the  $yz$  plane coincides with the plane of the vectors  $L$  and  $k$ , the  $x$  axis is perpendicular to the latter). The rotation of the  $xyz$  system relative to the  $x_0 y_0 z_0$  system is determined by the rotation of the plane of the director (the plane of the vectors  $L$  and  $k$ ), i.e., by the  $z$ -dependence of the azimuthal angle  $\psi$  between the axes  $y$  and  $y_0$ . For a uniaxial cholesteric with constant  $g$ , the derivative  $d\psi/dz$  also remains constant.

It is known that the dielectric tensor  $\epsilon_{ij}(\omega, r)$  of a uniaxial anisotropic medium without absorption is given by (Ref. 1, § 6.1)

$$\epsilon_{ij} = \epsilon_{\perp} \delta_{ij} + (\epsilon_{\parallel} - \epsilon_{\perp}) L_i L_j \quad (2.1)$$

Here  $L_i$  and  $L_j$  are the components of a unit vector  $L$  directed along the anisotropy axis,  $\delta_{ij}$  is the Kronecker symbol, and  $\epsilon_{\perp}$  and  $\epsilon_{\parallel}$  determine the squares of the refractive indices  $n_{\perp}^2$  and  $n_{\parallel}^2$  of the ordinary and extraordinary waves with wave vector  $k \perp L$ . In a liquid crystal with a spatially varying orientation of the director  $L$ , the tensor  $\epsilon_{ij}(\omega, r)$  retains the form (2.1). If it is considered in the local coordinate system  $xyz$  ( $z \parallel k$ ), then  $L_x = 0$  as a result of the foregoing choice of the axes  $x$  and  $y$ . It is also convenient to write down the electromagnetic-field equations in terms of projections on the axes  $x$ ,  $y$ , and  $z$ , which rotate together with the optical axes of the crystal. In a homogeneous cholesteric (with uniform director rotation  $d\psi/dz = 2\pi/g_0 = \text{const}$ ), these reduce to a system of linear differential equations with

constant coefficients. Their solutions define normal (helical) waves in the considered medium; for two helical waves propagating in the  $+z$  direction, the refractive indices  $\bar{n}_{1,2}$  and the polarization coefficients

$$\bar{K}_{1,2} = -iE_y^{(1),(2)}/E_x^{(1),(2)} \quad (2.1)$$

are given respectively by<sup>18</sup>

$$\bar{n}_{1,2} = \varepsilon^{\pm} \pm \psi' (1 + \bar{q}^2)^{\pm 1/2}; \quad \bar{K}_{1,2} = \bar{q} \mp (1 + \bar{q}^2)^{\pm 1/2}. \quad (2.2)$$

The index 1 and the upper signs in (2.2) pertain to a wave of one type, while the index 2 and the lower signs to the wave of the other type; the parameter is

$$\bar{q} = (n_o - n_e) / 2\psi' \approx \omega \Delta \varepsilon \sin^2 \alpha / 4c\varepsilon^{\pm} d\psi/dz; \quad (2.3)$$

the derivative is  $\psi' = d\psi/d\zeta$ , where  $\zeta = \omega z/c$  is the dimensionless coordinate; the quantity  $\varepsilon^{\pm 1/2}$  plays the role of the average refractive index:

$$\varepsilon^{\pm} = (n_o \pm n_e) / 2 \approx [\varepsilon_{\parallel}^{\pm} + (\varepsilon_{\perp} + \Delta \varepsilon \cos^2 \alpha)^{\pm 1/2}] (1 + \Delta \varepsilon \cos^2 \alpha / \varepsilon_{\perp})^{\pm 1/2}. \quad (2.4)$$

In formulas (2.3) and (2.4) we have  $\Delta \varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$ , and the refractive indices  $n_o$  and  $n_e$  of the ordinary and extraordinary waves are connected in the following manner with the local values of the dielectric tensor (2.1):

$$n_o = \varepsilon_{\perp}^{\pm}, \quad n_e = [\varepsilon_{\parallel} \varepsilon_{\perp} / (\varepsilon_{\perp} \sin^2 \alpha + \varepsilon_{\parallel} \cos^2 \alpha)]^{\pm 1/2}. \quad (2.5)$$

Here  $\alpha$  is the angle between the director  $\mathbf{L}$  and the light propagation direction  $\mathbf{k}$ . The refractive indices  $n_o$  and  $n_e$  must be distinguished from the refractive indices  $\bar{n}_1$  and  $\bar{n}_2$  of the helical waves (2.2). The helical waves are identical with the ordinary and extraordinary waves only in the limit of infinitely large helix pitch  $g \rightarrow \infty$  [i. e.,  $\psi' \rightarrow 0$ , see formula (2.2)].

It follows from (2.2) that the helical waves of orthogonal polarizations:  $\bar{K}_2 = -\bar{K}_1^{-1}$ . At  $\bar{q}^2 \gg 1$ , the polarization is close to linear, and at  $\bar{q}^2 \ll 1$  it is close to circular with an electric-field vector rotating in the opposite direction.

The explicit expressions (2.2) for  $\bar{n}_{1,2}$  and  $\bar{K}_{1,2}$  are valid in the region

$$|\bar{n}_1 - \bar{n}_2| \ll \bar{n}_1 + \bar{n}_2, \quad (2.6)$$

where the dispersion branches of the helical waves are close. These expressions are quite adequate for the subsequent analysis, inasmuch as in a smoothly inhomogeneous medium ( $\Lambda \gg \lambda = 2\pi\sqrt{\varepsilon} k_0$ ) the geometrical-optics violation accompanied by the effect of the linear interaction can take place only in the region where the dispersion branches come close together (see below). In addition, the inequality (2.6) allows us to neglect the reflected waves and to consider only the interaction of two co-moving helical waves (cf.<sup>20,21</sup>).

In a smoothly inhomogeneous liquid crystal with values of  $\varepsilon_{\parallel}$ ,  $\varepsilon_{\perp}$ , and  $\psi'$  that vary along the  $z$  axis over a scale  $\Lambda$  satisfying the condition

$$k_0 |\bar{n}_1 - \bar{n}_2| \Lambda \gg 1, \quad (2.7)$$

it is legitimate to use the asymptotic (geometrical-optics) approximation for high-frequency electromagnetic fields. This approximation describes a superposition of helical waves, and the electric field of these waves, which propagate in the same direction ( $+z$ ), is given by

$$E_x = f_1 [\bar{n}_1 (1 + \bar{K}_1^2)]^{-1/2} + f_2 [\bar{n}_2 (1 + \bar{K}_2^2)]^{-1/2}, \quad (2.8)$$

$$E_y = i\bar{K}_1 f_1 [\bar{n}_1 (1 + \bar{K}_1^2)]^{-1/2} + i\bar{K}_2 f_2 [\bar{n}_2 (1 + \bar{K}_2^2)]^{-1/2},$$

where

$$f_{1,2} = A_{1,2} \exp \left( -i \int \bar{n}_{1,2} d\zeta \right), \quad A_{1,2} = \text{const}_{1,2}. \quad (2.9)$$

We emphasize that the geometrical optics of helical waves takes into account, in addition to the inhomogeneity of the quantities  $\varepsilon_{\parallel}$  and  $\varepsilon_{\perp}$ , also the non-uniformity of the rotation of the director in a weakly inhomogeneous cholesteric. The definition of the corresponding inhomogeneity scale  $\Lambda \sim \psi' / k_0 \psi''$  contains therefore the second derivative of  $\psi$ . If there is no rotation of the director  $\mathbf{L}$  (when  $\psi' = 0$ ) the geometrical optics of the helical waves reduces to the geometrical optics of ordinary and extraordinary waves in the usual uniaxial crystal.

In regions where the inequality (2.7) is violated, the high-frequency field cannot be represented as a superposition of the helical waves (2.8) and (2.9), despite the smoothness of the inhomogeneity of the medium over the wavelength (but not over the period of the spatial beats of the geometrical-optics wave). In this case the functions  $f_{1,2}$  in (2.8) cannot be expressed in the form (2.9). It can be shown, however, by regarding (2.8) as Maxwell's equations with  $E_x$  and  $E_y$  replaced by the variables  $f_1$  and  $f_2$ , that the latter are defined by the system of coupled equations

$$f_1' + i\bar{n}_1 f_1 = -\bar{\Psi} f_2, \quad f_2' + i\bar{n}_2 f_2 = \bar{\Psi} f_1; \quad (2.10)$$

$$\bar{\Psi} = -\bar{q}' / 2(1 + \bar{q}^2)$$

(the primes denote differentiation with respect to the dimensionless variable  $\zeta = k_0 z$ ). In the theory of radio wave propagation in the plasma, such a system is known as the Budden equations.<sup>22</sup> In the geometrical optics approximation (2.7), the solution of the system (2.10) reduces to the form (2.9).

The system (2.10) is valid under the condition (2.6), which allows us to neglect the reflected waves. According to (2.2), it is equivalent to simultaneous satisfaction of the following two inequalities:

$$(\psi')^2 \ll \varepsilon; \quad |n_o - n_e| \ll n_o + n_e. \quad (2.11)$$

The first of them means that the helix pitch<sup>1)</sup>  $g \gg \lambda$ . Usually  $g \sim 1-10 \mu\text{m}$ , i. e., it is comparable with or larger than the light wavelength  $\lambda$ . In a pure cholesteric,  $g$  can be increased to values that ensure satisfaction of the strong inequality  $g \gg \lambda$  for the optical and infrared bands by changing the external conditions, while in a nematic with admixture of chiral molecules that induce helical ordering of all the molecules this can be done by varying its chemical composition (Ref. 1, § 6.2, Ref. 2, § 6.1, Ref. 23). The second inequality in (2.11) is equivalent to the requirement of low anisotropy of the liquid crystal  $|\varepsilon_{\parallel} - \varepsilon_{\perp}| \ll \varepsilon$  [see (2.1) and (2.5)]. Usually this condition is satisfied, with  $|\varepsilon_{\parallel} - \varepsilon_{\perp}| \leq 0.1\varepsilon$ . We note that the weakness of the anisotropy allows us to extend the region of applicability of the system (2.10) to include the case of three-dimensionally inhomogeneous liquid crystals of the cholesteric type. In this case the system (2.10) describes the interaction of helical waves in the case of light propagation along a "quasi-isotropic" ray whose shape is determined by the

eikonal equation in an isotropic medium with a refractive index  $(n_o + n_e)/2$ . This "quasi-isotropic approximation" is vitally used to investigate wave interaction in a magnetoactive plasma (see Refs. 24, 5, 17).

As noted in the introduction, the effective linear transformation, which occurs when waves pass through the region in which the geometrical optics is violated (1.1), consists primarily of a change in the ratio of the amplitudes of the helical waves (i. e., of the quantity  $f_1/f_2$ ). To investigate this effect it is therefore expedient to change from the system (2.10) directly to the equation for the function  $P = -if_1/f_2$ :

$$dP/d\eta = i(P^2 - 1) - 2iG(\eta)P. \quad (2.12)$$

Recognizing that, according to (2.10),

$$|f_1(\zeta)|^2 + |f_2(\zeta)|^2 = \text{const}$$

(the law of energy conservation along the ray). In the Riccati equation (2.12), the independent variable

$$\eta = \frac{\pi}{4} + \frac{1}{2} \text{arctg } \tilde{q} \quad (0 \leq \eta \leq \pi/2) \quad (2.13)$$

characterizes the polarization coefficients of the helical waves:

$$\text{tg } \eta = \tilde{K}_2 = -\tilde{K}_1^{-1}.$$

At  $\eta$  close to zero or  $\pi/2$ , the polarization is close to linear, while at  $\eta \approx \pi/4$  it is close to circular. The entire information on the interaction is contained in the function

$$G(\eta) = (\tilde{n}_2 - \tilde{n}_1) / 2\tilde{\Psi} = 2\psi'(1 + \tilde{q}^2)^{1/2} / \tilde{q}', \quad (2.14)$$

which is determined by the cholesteric-helix pitch  $g(\zeta) = 2\pi/k_0\psi'$  and by the form of the function  $q(\zeta)$  (2.3), which characterizes the change of the polarization of the helical waves along the ray [see (2.2)].

### §3. QUALITATIVE PICTURE OF LINEAR CONVERSION OF HELICAL WAVES

The effectiveness of the interaction on passage of the radiation through the crystal can be characterized by the conversion coefficient  $Q$  ( $0 \leq Q \leq 1$ ). It determines the relative intensity of the helical wave of one type as it emerges from the crystal, if a wave of another type enters the crystal:

$$Q = |f_1^{\text{out}}|^2 / |f_2^{\text{in}}|^2, \quad f_1^{\text{in}} = 0 \quad (3.1)$$

or

$$Q = |f_2^{\text{out}}|^2 / |f_1^{\text{in}}|^2, \quad f_2^{\text{in}} = 0. \quad (3.1a)$$

The conversion coefficient  $Q$  is connected with  $P(\eta)$  at the exit by the relation (for details see Ref. 17)

$$Q / (1 - Q) = |P(\eta_{\text{out}})|^2, \quad (3.2)$$

where  $P(\eta_{\text{out}})$  is the result of the solution of Eq. (2.12) under the boundary condition  $P(\eta_{\text{in}}) = 0$ , i. e.,  $f_1^{\text{in}} = 0$  [here  $\eta_{\text{in}}$  and  $\eta_{\text{out}}$  are the values of the variable  $\eta$  (2.13) at the entrance and exit from the crystal, respectively].

The values of the coefficient  $Q$  depend essentially on the character of the function  $G(\eta)$  and on the interval of variation of  $\eta$  in the crystal. The situation here is fully analogous with the propagation of electromagnetic waves

in a magnetoactive plasma. We shall therefore report below briefly the results of a qualitative analysis of the interaction, and refer the reader for more details to our earlier papers.<sup>5,17</sup> If  $|G(\eta)| \gg 1$  along the ray, then the geometrical-optics approximation is valid everywhere and  $Q \ll 1$  (the interaction is weak), and the helical waves propagate in the liquid crystal practically independently. On the other hand  $Q \ll 1$  regardless of the form of the function  $G(\eta)$  if the interval of variation of  $\eta$  along the ray is small:  $|\Delta\eta| \ll 1$ . If the liquid crystal is a "transition layer," meaning a medium with monotonic variation of the parameter  $\tilde{q}$  (2.3) that determines the character of the polarization  $\tilde{K}_{1,2}$  (2.2) of the helical waves, then under the condition  $|G(\eta)| \ll 1$  we have

$$Q = \sin^2 \Delta\eta = \sin^2 \left( \frac{1}{2} \text{arctg } \tilde{q}_{\text{out}} - \frac{1}{2} \text{arctg } \tilde{q}_{\text{in}} \right). \quad (3.3)$$

The interaction is determined in this case only by the difference between the polarization coefficients  $\tilde{K}_{1,2}(\tilde{q})$  of the helical waves at the exit from the crystal. In the general case, an effective interaction (with  $Q \sim 1$ ) is realized in the crystal under the condition that the function  $|G(\eta)| \leq 1$  over intervals  $|\Delta\eta| \sim 1$ . The last requirement means that there should exist in the liquid crystal a region in which

$$|\Delta\tilde{q}| \sim 1 \quad \text{at} \quad \tilde{q}^2 \sim 1 \quad (3.4)$$

[see (2.13)]. If the function  $G(\eta)$  (2.14) does not vary too strongly over the interval  $|\Delta\eta| \sim 1$  ( $|\Delta G| \leq |G|$ ), then the degree of transformation of the helical waves can be conveniently characterized by the "interaction parameter"

$$G = |G(\eta)|_{\tilde{q}^2 \sim 1} = |4\sqrt{2}\psi' / \tilde{q}'|_{\tilde{q}^2 \sim 1}, \quad (3.5)$$

which is the value of the function  $G(\eta)$  in the region where the polarization of the helical waves is essentially elliptic. When  $G \gg 1$ , the interaction is weak ( $Q \ll 1$ ); at  $G \sim 1$  the coefficient  $Q$  is comparable with unity;  $Q$  becomes maximal for values  $G \ll 1$  [see (3.3)].

Under concrete conditions of inhomogeneous liquid crystals, there is no wave transformation if the orientation of the plane of the director  $L$  remains unchanged ( $\psi' = 0$ ). In fact, in this case we have  $\tilde{q}^2 = \infty$ ,  $\Delta\eta = 0$ , and  $Q = 0$  [see (2.3), (2.13), (3.3)]. The onset of linear interaction can be due in this case only to the natural or magnetic optical activity of the crystals, i. e., to off-diagonal dielectric-tensor components in (2.1), which are usually very small. In this respect, liquid crystals differ substantially from a plasma in a magnetic field, in which effective wave interaction is possible not only on account of a change in the orientation of the magnetic field, but also as a result of strong gyrotropy of the medium (see Refs. 5 and 16). It follows from the foregoing that in inhomogeneous liquid crystals, independently of the condition (2.7), the geometrical-optics approximation can be used to describe any effect that is not connected with the helical structure (e. g., the  $S$  and  $B$  effects, concerning which see Chap. 4 of Ref. 2). At the same time, for a consistent description of twisted nematics (and the  $T$ -effect in them), of cholesterics, and of smectics of type  $C$  with admixture of chiral molecules, it is essential to take into account the linear in-

teraction of the helical waves. However, even for these objects, the wave conversion is small if the twist of the helix is weak:

$$2|\psi'| \ll |n_e - n_o|. \quad (3.6)$$

The polarization of the helical waves is then close to linear [ $\tilde{q}^2 \gg 1$ , see (2.2) and (2.3)] and varies slowly along the ray, thus ensuring smallness of the interval  $|\Delta\eta| \ll 1$ .

The linear interaction remains insignificant also in the case opposite to (3.6) (strongly twisted helix). In this case the helical waves will be circularly polarized ( $\tilde{q}^2 \ll 1$ ) and again  $|\Delta\eta| \ll 1$ . A strong transformation of helical waves can be realized in liquid crystals, where a transition takes place from a strongly twisted ( $\tilde{q}^2 \ll 1$ ) to a weakly twisted ( $\tilde{q}^2 \gg 1$ ) helix through a region in which  $\tilde{q}^2 \sim 1$  ["transition layer," see (3.4)]. In this region, the polarization of the helical waves is elliptic, and the pitch of the cholesteric helix  $g = 2\pi/k_0\psi'$  is comparable with the period  $2\pi/k_0|\tilde{n}_1 - \tilde{n}_2|$  of the spatial beats between the waves.

If the pitch of the helix [more accurately, the quantity  $\tilde{q}$  (2.3)] changes substantially in this transition, or in other words, if the scale  $\Lambda$  over which the polarization of the helical waves changes is less than or of the order of  $2\pi/k_0|\tilde{n}_1 - \tilde{n}_2|$  [the condition (1.1)], then the parameter  $G$  becomes less than or of the order of unity [see (2.14) and (3.5)]. In accord with the statements made above, the latter ensures transformation of helical waves with efficiency  $Q \sim 1$ .

#### §4. SELECTIVE PASSAGE OF LIGHT IN A CRYSTAL WITH AN INHOMOGENEOUS CHOLESTERIC HELIX

We consider a liquid crystal in which the helix pitch varies along the direction of the light ray monotonically like

$$\psi' = \psi''\zeta, \quad \psi'' = \text{const} > 0 \quad (4.1)$$

(linear layer). The importance of this problem lies in the possibility of approximating smooth functions  $\psi'(\zeta)$  on individual sections in an inhomogeneous cholesteric by means of the relation (4.1). In the case (4.1) the parameter is

$$\tilde{q}(\zeta) = (n_e - n_o)/2\psi''\zeta.$$

The function  $G(\eta)$  (2.14) takes the form [we assume that  $\alpha$  is constant, see (2.5)]

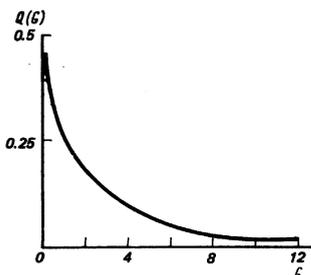


FIG. 1. Dependence of the conversion coefficient  $Q(G)$  (4.4) on the interaction parameter  $G$ ;  $Q(G) \approx 1/2 - (\pi G/32\sqrt{2})^{1/2}$  at  $\tilde{q}_{in}^{-2} \ll G \ll 1$ , and  $Q(G) \approx 2G^{-2}$  at  $1 \ll G \ll \tilde{q}_{out}^{-2}$ .

$$G(\eta) = \frac{(n_e - n_o)^2}{2\psi''} \cos^{-3} 2\eta, \quad (4.2)$$

and Eq. (2.12) has an analytic solution (see Ref. 25).

Let the crystal layer be located at  $\zeta < 0$ , and then, according to (4.1), a transition takes place in the layer from a weakly twisted into a strongly twisted helix:  $\tilde{q}_{in}^2 \gg 1$ ,  $\tilde{q}_{out}^2 \ll 1$ . Using the asymptotic forms of the exact solution at  $G\tilde{q}_{in}^2 \gg 1$  and  $G\tilde{q}_{out}^2 \ll 1$ , where

$$G = |G(\eta)|_{\tilde{q}^2 \rightarrow 1} = \sqrt{2}(n_e - n_o)^2/\psi'', \quad (4.3)$$

we obtain the following expression for the conversion coefficient in the layer:

$$Q = \left[ 1 + \left| \frac{1+w(G)}{1-w(G)} \right|^2 \right]^{-1}, \quad (4.4)$$

$$w(G) = \frac{\sqrt{G}\Gamma^2(-iG/16\sqrt{2})}{2^{1/4}\sqrt{\pi}\Gamma(-iG/8\sqrt{2})} \exp\left(-i\frac{\pi}{4} - iG\frac{\ln 2}{8\sqrt{2}}\right)$$

( $\Gamma$  is the gamma function). It is clear from (4.4) (see Fig. 1) that with increasing  $G$  the value of  $Q$  decreases monotonically from  $Q = 1/2$  (at  $G = 0$ ) to  $Q = 0$  (as  $G \rightarrow \infty$ ).<sup>2)</sup> In the region  $1 \ll G \ll \tilde{q}_{out}^{-2}$  the conversion coefficient is  $Q \approx 2G^{-2}$ . In accordance with the qualitative picture of §3, the conversion of the helical waves becomes substantial ( $Q \sim 1$ ) if the interaction parameter  $G \lesssim 1$ .<sup>3)</sup>

If light containing one helical wave (linearly polarized in the plane of the director) is incident on such a cell, then the radiation leaving the cell contains helical waves that are coherent with each other and have right- and left-hand circular polarization. The resultant polarization of the light becomes elliptic with a ratio of the principal axes

$$(-\sqrt{Q} + \sqrt{1-Q})/(\sqrt{Q} + \sqrt{1-Q})^{-1}.$$

The light remains linearly polarized in the case of strong interaction ( $G \ll 1$ ,  $Q \approx 1/2$ ), and is circularly polarized if  $G \gg 1$  and  $Q \approx 0$ .

In the problem considered here, the parameter  $G$  (4.3) has a rather strong frequency dependence,  $G(\omega) \propto \omega^4$ . The reason is that for estimates in liquid crystals one can put  $n_e(\omega) - n_o(\omega) \propto \omega$  (see Ref. 26), and

$$\psi'' = (c^2/\omega^2) d^2\psi/dz^2 \propto \omega^{-2}.$$

This dependence leads to a strong decrease of the conversion coefficient  $Q$ , from  $Q_{max}$  to  $Q \ll 1$ , with increasing frequency in a relatively narrow frequency interval near those values of the frequency  $\omega_0$  at which  $G(\omega_0) = 1$ . Estimates show that  $\Delta\omega/\omega_0 \geq 0.1$  for liquid crystals.

The nonlinear interaction makes it possible, e.g., to realize a filter that transmits radiation only at low frequencies  $\omega < \omega_0$ . To this end, a liquid-crystal cell with non-uniform helix of the type (4.1) must be placed between two polarizers. If the first polarizer transmits radiation with polarization corresponding to that of one of the helical waves at the entrance to the cell, and the second polarizer transmits only radiation corresponding to the second helical wave at the exit from the cell, then at high frequencies  $\omega > \omega_0$ , where there is no effective conversion of one wave into another, the radiation does not pass through the filter. At low frequen-

cies  $\omega < \omega_0$  the filter is partially transparent—it transmits the radiation corresponding to polarization of the helical wave produced as a result of linear conversion in the interaction region.

## §5. PASSAGE OF LIGHT THROUGH A LIQUID CRYSTAL WITH UNTWISTED HELIX

In liquid crystals, the form of the cholesteric helix changes substantially when an electric field  $E$  is applied (an external magnetic field acts similarly). If this field is uniform and is oriented perpendicular to the helix axis  $z$ , then its influence manifests itself primarily on the helix pitch  $g$ . With increasing field intensity  $E$ , the rotation of the director  $L$  becomes non-uniform; the  $\psi(z)$  dependence in the free cholesteric helix is then described by the formula

$$\psi(z) = \text{Arc sin sn} \left[ \frac{z}{\kappa \xi}, \kappa \right], \quad (5.1)$$

where the so-called coherence length  $\xi$  and the modulus of the elliptic cone  $\kappa$  ( $0 \leq \kappa \leq 1$ ) are determined by the parameters of the cholesteric and by the magnitude of applied field  $E$  (Ref. 1, § 6.2, Ref. 2, § 6.5). We assume for simplicity that the director  $L$  is everywhere orthogonal to the helix axis  $z$ . It is clear from (5.1) that as  $E$  approaches the critical value  $E_{cr}$  determined by the properties of the cholesteric (when  $\kappa$  increases and approaches unity), the pitch of the helix increases, and the deformed helix itself looks like a sequence of rather abrupt rotations of the director through an angle  $\psi_0 = \pi$ , separated by layers of practically constant orientation of  $L$  (see also § 10, Fig. 4). The distance  $l$  between the reversal regions, i. e., the characteristic dimension of the "domain" in which  $L \parallel E$ , depends logarithmically on the difference between  $E$  and the critical field  $E_{cr}$  (see Ref. 2, § 6.5):

$$l = \frac{g_0}{\pi^2} \ln \left[ \frac{16E}{E_{cr} - E} \ln \left( \frac{4\sqrt{e}E}{E_{cr} - E} \right) \right] \quad (5.2)$$

(if  $E_{cr} - E \ll E_{cr}$ ). The abrupt rotation of the director through an angle  $\pi$  ("domain wall," see Fig. 2) is described by the relation

$$\psi = \frac{2\psi_0}{\pi} \text{arctg exp} \left( \frac{z}{\xi} \right) - \frac{\psi_0}{2} \quad (-\infty < z < +\infty), \quad (5.3)$$

which follows from (5.1) in the limit as  $E \rightarrow E_{cr}$  ( $\kappa \rightarrow 1$  and  $\xi \rightarrow g_0/\pi^2$ ). Indeed, representing (5.1) in the form

$$\frac{z}{\xi \kappa} = \int_0^{\psi(z)} \frac{d\varphi}{(1 - \kappa^2 \sin^2 \varphi)^{1/2}},$$

we obtain as  $\kappa \rightarrow 1$

$$\frac{z}{\xi} = \ln \text{tg} \left( \frac{\psi}{2} + \frac{\pi}{4} \right),$$

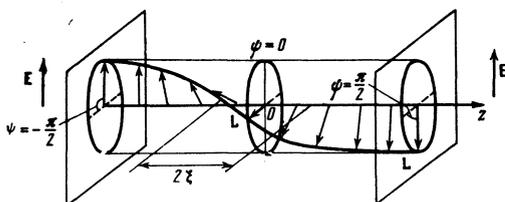


FIG. 2. Spatial distribution of director  $L(z)$  (5.3) in an untwisted cholesteric ("domain wall").

which is equivalent to (5.3) at  $\psi_0 = \pi$ . The characteristic scale of the region of rotation of  $\xi = g_0/\pi^2$  is smaller by a factor  $\pi^2$  than the unperturbed pitch of the helix  $g_0$ . Starting with the critical field  $E = E_{cr}$  (the cholesteric-nematic transition field), the formation of domain walls is not energetically favored and there is no helix [ $l \rightarrow \infty$ , see (5.2)].

The characteristic function  $G(\eta)$  (2.14) corresponding to (5.3) takes the form

$$G(\eta) = \pm \kappa_0 (n_s - n_o) \xi \cos^{-2} 2\eta \sin^{-1} 2\eta \left\{ 1 - \left[ \frac{\pi \kappa_0 (n_s - n_o) \xi}{2\psi_0} \right]^2 \text{tg}^2 2\eta \right\}^{-1/4},$$

where the plus sign corresponds to the first half of the layer ( $-\infty < \zeta < 0$ ), and the minus sign to the second half ( $0 < \zeta < \infty$ ). The solution of the problem of passage of waves through a cholesteric with a helix of the type (5.3) is known: in this case Eq. (2.2) has an exact solution<sup>5</sup> at any  $\psi_0$ , so that we can determine the transformation coefficient

$$Q = \sin^2 \psi_0 / \text{ch}^2 [\pi \kappa_0 (n_s - n_o) \xi / 2]. \quad (5.4)$$

It is clear therefore that for a strongly deformed helix with one rotation through  $\psi_0 = \pi$  (and consequently also for a cell with an integer number of such rotations) there is no linear transformation even in the case of abrupt rotations over a small scale  $\xi$ . This result is connected with the symmetry of the layer (5.3), on both sides of which the properties of the crystal are perfectly identical (rotation of the director through an angle  $\pi$  returns the crystal to its previous position with the same orientation of the anisotropy axis; the last statement does not pertain to the case of ferroelectric smectics, see Ref. 27). The layer (5.3) consists in fact of two transition layers separated by a plane  $\zeta = 0$ ; the linear-interaction effects that arise in these layers at  $G \leq 1$  cancel each other.

In a crystal whose structure can be approximated by the function (5.3) with  $\psi_0 \neq \pi m$  ( $m$  is an integer), the conversion coefficient  $Q$  (5.4) becomes different from zero. A cell of this type, in which the director is rotated through an angle  $\psi_0 = \pi/2$ , can apparently be realized by placing the cholesteric between two crossed polarizers in a specially chosen inhomogeneous electric field. According to (5.4), the effective conversion will take place if the thickness of the transition layer is

$$\xi \leq \xi_{cr} = |\pi \kappa_0 (n_s - n_o) / 2|^{-1}.$$

The propagation of the light in the cell obeys geometrical optics if  $\xi \gg \xi_{cr}$ . If the polarizers located at the ends of the cell transmit helical waves polarized linearly in the plane of the director  $L$ , then the light will pass through the cell in the case  $\xi \gg \xi_{cr}$  (open cell).

On the other hand if  $\xi \ll \xi_{cr}$ , then the cell is closed: under conditions of strong interaction, the linearly polarized light emerging from the first polarizer propagates in the crystal without change of the plane of polarization (relative to the laboratory coordinate frame  $x_0 y_0 z$ ); it will therefore be blocked by the second polarizer, whose plane is perpendicular to the plane of polarization of the light, since  $\psi_0 = \pi/2$ . The described system can obviously be used as a light shutter. The change of the inhomogeneity scale  $\xi$  required for this

purpose (by changing the external field  $\mathbf{E}$ ) can be effected within relatively narrow limits, since  $\xi$  is contained in (5.4) in the argument of a hyperbolic cosine. This circumstance decreases the inertia of such a device.

## §6. ORIENTATIONAL PHASE TRANSITION AND POLARIZATION CUTOFF OF LIGHT IN LIQUID CRYSTALS

A change in the external electric or magnetic field, as already mentioned in the introduction, can lead to second-order orientational phase transitions (revealed, e.g., by a change in the capacitance<sup>14,28</sup>), and consequently to a change of the optical properties of a liquid-crystal structure, owing to the linear interaction of the light waves. As a result, the polarization properties of a liquid-crystal structure and, in particular, the optical threshold of the polarization cutoff of the light, turn out to be connected with the molecular properties of the liquid crystal and with the critical phase-transition point. A study of the optical threshold is therefore of substantial interest for the physics of the condensed-liquid-crystal state of matter.

To observe the polarization light cutoff actually produced by linear conversion of the waves, polarizers placed on both sides of the liquid-crystal layer pass only one of the helical waves. Changing the external field beyond the critical point of the phase transition leads to a deformation of the twist structure, and hence to an increase of the drop  $\Delta\tilde{q}$  and of the derivative  $d\tilde{q}/d\psi$  [see (3.4) and (3.5)], i.e., to an enhancement of the linear interaction between the helical waves incident on the layer and emerging from the layer. A characteristic optical transition thus sets in after the phase transition. The optical transition can be quite abrupt, and we shall speak, to be specific, of a polarization light-cutoff threshold defined by the conditions  $|\Delta\tilde{q}| \sim 1$  and  $G \sim 1$  at  $\tilde{q}^2 \sim 1$  (see §3). The width of the optical threshold can, e.g., be characterized by that change of field at which the values of  $\Delta\tilde{q}$  and  $G$  are doubled.

Most experiments on polarization cutoff of light were performed on planar twisted structures<sup>7,14,15,28-31</sup> of thickness  $d \sim 0.01-1$  mm, formed between two parallel glass plates; their inert surfaces are so finished that the long axes of the molecules (the director  $\mathbf{L}$ ) are oriented along the walls (see Ref. 32 concerning the methods of the orientation). If such a twist structure is uniform, then, on moving along the helix axis  $z$ , the polarization ellipse of the light corresponding to the helical wave of one type is rotated and follows strictly the azimuthal rotation of the director  $\mathbf{L}$ . Therefore the polarization ellipse of the light emerging from the twist structure is turned relative to the initial polarization ellipse through an angle  $h\delta + \psi_0$  ( $h = 2\pi/g_0$  is the wave number of the "free" helix and  $\psi_0$  is the angle of the forced twisting of the helix<sup>4</sup>). This geometrical-optics regime has been well established<sup>33</sup> and was in fact known already to Mauguin.<sup>34</sup> This regime can be upset only in an inhomogeneous helix, when external fields exceeding the critical deformation field are applied to the twist structure. This is confirmed by direct observations<sup>7,14</sup> and by numerical experiments.<sup>28</sup>

It is clear from the foregoing that to determine the optical threshold and its connection with the critical field of the phase transition it is necessary to know how the liquid-crystal structure is deformed; according to the exposition in §3, the structure must definitely be helical (twist structure) in order for an optical threshold to exist.

According to the continual theory of nematics and cholesterics,<sup>1</sup> under specified boundary conditions (rigid coupling of the molecules with the walls) a helical structure with minimum free energy is established in the liquid crystal by the action of the external field. The volume density of the free energy is then written in the form

$$\Phi = \frac{K_1}{2}(\text{div } \mathbf{L})^2 + \frac{K_2}{2}(\mathbf{L} \cdot \text{rot } \mathbf{L} + h)^2 + \frac{K_3}{2}[(\mathbf{L} \cdot \nabla) \mathbf{L}]^2 - \frac{\mathbf{D} \cdot \mathbf{E}}{8\pi}, \quad (6.1)$$

where  $\mathbf{E}$  is the static external electric field,  $\mathbf{D} = \epsilon_{L0}\mathbf{E} + \Delta\epsilon_0(\mathbf{L} \cdot \mathbf{E})\mathbf{L}$  is its induction;  $\Delta\epsilon_0 = \epsilon_{10} - \epsilon_{L0}$ ;  $\epsilon_{10}$  and  $\epsilon_{L0}$  are the static dielectric constants measured along and across  $L$ , respectively.<sup>5</sup> The elastic constants  $K_1$ ,  $K_2$ , and  $K_3$  in (6.1) pertain respectively to the transverse flexure, torsion, and longitudinal flexure.

It is easy to prove that in a liquid-crystal structure a phase transition is possible only from a state in which the director  $\mathbf{L}$  is everywhere directed along or across the external field  $\mathbf{E}$ .<sup>2,35</sup> Otherwise the reorientation of the director  $\mathbf{L}$  with changing electric field proceeds smoothly (without jumps of the derivatives), and the critical field of the phase transition can be defined only arbitrarily by connecting it with the steepest section of the plot of the conformation against the field. (The vanishing of the phase transition is not connected with the non-rigid adhesion of the molecules to the walls,<sup>36</sup> but is due to the appearance of the torque

$$\frac{1}{4\pi}[(\mathbf{D} - \mathbf{E}) \times \mathbf{E}] = \frac{\Delta\epsilon_0}{4\pi}(\mathbf{L} \cdot \mathbf{E})[\mathbf{L} \times \mathbf{E}],$$

that acts on the electric dipoles (Ref. 1, §3.2). Since the optical threshold exists only in helical structures, the only configuration of the cholesteric type in which it is possible to observe both polarization cutoff of the light and a phase transition is a twist structure in a field directed along the helix axis (see §§7, 8).

There is another variant, wherein there is no helical structure in the initial state and the condition  $\mathbf{L} \cdot \mathbf{E} = 0$  or  $\mathbf{L} \times \mathbf{E} = 0$  is satisfied. If the phase transition leads to formation of a structure of the cholesteric type, then it can cause also polarization cutoff of the light (see below).

In a planar twist structure, the second-order phase transition is the result of the fact that the electric field effectively tilts the molecules towards the axis of the helix (at  $\Delta\epsilon_0 > 0$ ). The phase-transition critical field  $E_{cr}$  at which the angle  $\alpha$  between the director  $\mathbf{L}$  and the  $z$  axis of the helix begins to differ from  $\pi/2$  can be obtained from the relation

$$\frac{\Delta\epsilon_0 E_{cr}^2}{4\pi} = \left(\frac{\pi}{d}\right)^2 K_1 + \left(\frac{\psi_0}{d}\right)^2 (K_3 - 2K_2) + 2h \frac{\psi_0}{d} (K_3 - K_2) + h^2 K_3. \quad (6.2)$$

It is obtained by investigating the stability of a state

with a given free energy (6.1) by the de Gennes method (Ref. 1, §3.2.3). The first two terms in the right-hand side of (6.2) correspond to the nematic twist effect,<sup>31</sup> and the last two terms are due to the cholesteric properties of the liquid crystal.

For a field  $E$  exceeding the critical deformation field<sup>6)</sup>  $E_{cr}$ , the distribution of the director  $L(z)$  remains symmetrical about the cell center  $z=d/2$ , and is described by a system of Euler's equations:

$$\left(\frac{d\alpha}{dz}\right)^2 = \left(\frac{\Delta\epsilon_0 E^2}{4\pi K} - h^2\right) (\sin^2 \alpha - \sin^2 \alpha_0) + h_0^2 \sin^2 \alpha_0 \left(1 - \frac{\sin^2 \alpha_0}{\sin^2 \alpha}\right), \quad (6.3)$$

$$\frac{d\psi}{dz} = h + h_0 \frac{\sin^2 \alpha_0}{\sin^2 \alpha}. \quad (6.4)$$

This system is satisfied by the angles  $\alpha(z)$  and  $\psi(z)$ , which minimize the free energy (6.1). Here  $\alpha_0 = \alpha(d/2)$  is the minimum value that the angle  $\alpha$  reaches at the center of the layer; the constant

$$h_0 = \psi_0 / \int_0^d \frac{\sin^2 \alpha_0}{\sin^2 \alpha} dz$$

is connected with the azimuthal torsion of the twist structure. Equations (6.3) and (6.4) are written in an approximation in which

$$K = K_1 = K_2 = K_3, \quad (6.5)$$

and without allowance for the dependence of the field  $E$  on the coordinate  $z$ . This approximation does not influence the qualitative character of the results and makes it possible to simplify the exposition noticeably. There is no need to solve the system (6.3) and (6.4), since to determine the optical threshold it suffices to have information on the derivatives  $d\alpha/dz$  and  $d\psi/dz$ , as well as the value of the drop  $\Delta\tilde{q}$  in the region  $\tilde{q}^2 \sim 1$  (see §3). Calculating the integrals of (6.3) and (6.4) term by term over the thickness of the layer, and using the boundary conditions for a planar twist structure, we arrive at the following implicit equations for the layer parameters  $\alpha_0$  and  $h_0$ :

$$\begin{aligned} d \left( \frac{\Delta\epsilon_0 E^2}{4\pi K} - h^2 + h_0^2 \sin^2 \alpha_0 \right)^{1/2} &= 2F \left( \frac{\cos \alpha_0}{a} \right); \\ h_0 d \sin^2 \alpha_0 \Pi(\cos^2 \alpha_0, \cos \alpha_0/a) &= \psi_0 F(\cos \alpha_0/a), \end{aligned} \quad (6.6)$$

where

$$a = \left( 1 + \frac{h_0^2 \sin^2 \alpha_0}{\Delta\epsilon_0 E^2 / 4\pi K - h^2} \right)^{1/2},$$

$F$  and  $\Pi$  are complete elliptic integrals of the first and third kind, respectively.

According to (3.4), the threshold effect of the polarization cutoff of the light is strong if at the point of the phase transition (6.2), when  $d\psi/dz \equiv h + \psi_0/d$ , the following condition is valid

$$\tilde{q}_0 = \frac{\omega \Delta\epsilon}{4c\sqrt{\epsilon}} \left( \frac{\psi_0}{d} + h \right)^{-1} > 1 \quad (6.7)$$

(we assume for the sake of argument that  $\Delta\epsilon > 0$  and the helix is right-handed, i.e.,  $d\psi/dz > 0$ ). The inequality (6.7) is satisfied in practically all the experiments (see, e.g., Refs. 7 and 28), inasmuch as twist-structures with weak torsion are customarily used for the light cutoff. In this case the polarization cutoff of the light can take place only if the planar structure is suffi-

ciently strongly deformed, when  $\alpha_0 \lesssim q_0^{-1/2}$  at the center of the layer [the condition  $\tilde{q} \sim 1$ , see (2.3) and (3.4)]. Using this circumstance, we obtain from (6.6)

$$\begin{aligned} \alpha_0 &= 4 \cos \left( \frac{\psi_0}{2} \right) \exp \left[ -\frac{d}{2} \left( \frac{\Delta\epsilon_0 E^2}{4\pi K} - h^2 \right)^{1/2} \right], \\ h_0 &= \operatorname{tg} \left( \frac{\psi_0}{2} \right) \left( \frac{\Delta\epsilon_0 E^2}{4\pi K} - h^2 \right)^{1/2}. \end{aligned} \quad (6.8)$$

Knowing now the character of the deformation, we can write down explicit expressions for  $\tilde{q}$  and for the interaction parameter  $G$ ; these expressions determine the effectiveness of the conversion of the helical waves:

$$\tilde{q} = \tilde{q}_0 \frac{\sin^2 \alpha (h + \psi_0/d)}{h + h_0 \sin^2 \alpha_0 / \sin^2 \alpha}, \quad (6.9)$$

$$\begin{aligned} G &= 2\sqrt{2} (h \sin^2 \alpha + h_0 \sin^2 \alpha_0)^2 / \cos \alpha \sin \alpha (\sin^2 \alpha - \sin^2 \alpha_0)^{1/2} \\ &\times (h \sin^2 \alpha + 2h_0 \sin^2 \alpha_0) \left( \frac{\Delta\epsilon_0 E^2}{4\pi K} - h^2 + \frac{h_0^2 \sin^2 \alpha_0}{\sin^2 \alpha} \right)^{1/2} \tilde{q} \sim 1. \end{aligned} \quad (6.10)$$

In the last formula it is necessary to substitute the value of  $\sin \alpha$  determined from (6.9) under the condition  $\tilde{q} \sim 1$ . Depending on the relation between the pitch of the cholesteric helix and the thickness of the layer, this leads to different values of the interaction parameter  $G$ , and consequently to different light-cutoff regimes.

## §7. EFFECT OF CUTOFF OF THE MAUGUIN REGIME

In the nematic twist structure we have

$$h \ll (2\pi/d) \operatorname{tg}(\psi_0/2) \quad (7.1)$$

and the helices produced by twisting of the liquid crystal when the boundary plates are rotated through an angle  $\psi_0 \leq \pi/2$ . The inequality (6.7) is then satisfied: the thickness  $d$  of the planar structure is usually not less than 10  $\mu\text{m}$ . Addition of a small amount of cholesteric to lift the degeneracy with respect to the sign of the twist—the sign of the angle  $\psi$ —and to produce a homogeneous structure in the entire thickness of the layer<sup>31</sup> does not violate the inequalities (6.7) and (7.1). Moreover, according to (6.4) and (6.8), the value of  $\tilde{q}$  (2.3) at the walls of the layer

$$\tilde{q}_{in} = \tilde{q}_0 \psi_0 \exp \left[ d \left( \frac{\Delta\epsilon_0 E^2}{4\pi K} \right)^{1/2} \right] / 8d \sin \psi_0 \left( \frac{\Delta\epsilon_0 E^2}{4\pi K} \right)^{1/2},$$

increases rapidly with increasing field  $E$ . At the same time, at the center of the layer the quantity

$$\tilde{q}_{min} = \tilde{q}_0 \psi_0 \left( 4 \cos \frac{\psi_0}{2} \right)^2 \exp \left[ -d \left( \frac{\Delta\epsilon_0 E^2}{4\pi K} \right)^{1/2} \right] / d \operatorname{tg} \left( \frac{\psi_0}{2} \right) \left( \frac{\Delta\epsilon_0 E^2}{4\pi K} \right)^{1/2} \quad (7.2)$$

decreases exponentially. The reason for the latter is that practically the entire azimuthal rotation of the director through the angle  $\psi_0$  takes place at the central region of the layer, where the angle  $\alpha$  is small and the molecules are arranged almost along the  $z$  axis [see (6.4) at  $h=0$  and Fig. 3; this circumstance was not taken into account in the interpretation of the twist effect in Refs. 7 and in §4.4 of Ref. 2]. Taking the condition (3.4) into consideration, we see that the effective linear transformation of the helical waves (the cutoff of the Mauguin regime<sup>15,34</sup>) begins at  $\tilde{q}_{min} \leq 1/2$ . In this case the interaction parameter (6.10) is

$$G \approx \operatorname{tg}(\psi_0/2) (2\tilde{q}_{min})^{1/2} \ll 1$$

and decreases with increasing field

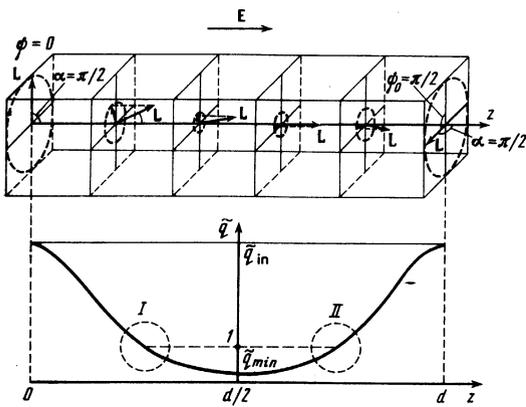


FIG. 3. Nematic twist structure in a longitudinal with a twist angle  $\psi_0 = \pi/2$  and characteristic form of the distribution of the quantity  $\bar{q} \propto \sin^4 \alpha$  (2.3) along the layer (the numbers I and II designate the regions of linear transformation of the helical waves).

$$E > E_{cr} = [4\pi K(\pi^2 - \psi_0^2 + h^2 d^2) / \Delta \epsilon_0 d^2]^{1/4}. \quad (7.3)$$

[Here  $E_{cr}$  is taken from (6.2) in the approximation (6.5).]

Thus, the optical threshold corresponding to the twist effect is obtained from the condition  $\bar{q}_{min} = 1/2$ , and is equal to

$$E_0 = E_{cr} \ln(\bar{q}^* / \ln \bar{q}^*) (\pi^2 - \psi_0^2)^{-1/4}, \quad (7.4)$$

$$\bar{q}^* = 2\bar{q}_0 \left(4 \cos \frac{\psi_0}{2}\right)^2 \psi_0 / \text{tg} \frac{\psi_0}{2}.$$

If  $\psi_0 = \pi/2$ , then  $\bar{q}^* = 8\pi\bar{q}_0$ ; in expression (6.7) for  $\bar{q}_0$  we must then put  $\sqrt{\epsilon} = \epsilon_0 \sqrt{\epsilon_1}$ , since the conversion of the light waves takes place in the region of small angles  $\alpha$  [see (2.4)]. It can be shown that if the strong inequality (7.1) is not satisfied, then the optical threshold for a liquid-crystal layer whose thickness does not exceed half the pitch of the free helix ( $h \lesssim \pi/d$ ) is defined by a similar relation

$$E_0 = E_{cr} \ln \left[ 2 \left(4 \cos \frac{\psi_0}{2}\right)^2 \bar{q}_0 \right] [\pi^2 - \psi_0^2 + h^2 d^2 (1 - E_{cr}^2 / E_0^2)]^{-1/4}. \quad (7.5)$$

It is clear from (7.4) and (7.5) that the optical threshold exceeds noticeably the critical field of the phase transition for thick liquid-crystal layers of nematics with large optical anisotropy  $\Delta \epsilon$ , which have a large  $\bar{q}_0$  (6.7). In practice, however, it is a complicated matter to obtain planar structures with  $\bar{q}_0 > 100$ ; the optical threshold  $E_0$  therefore differs usually from the critical field  $E_{cr}$  of the phase transition by approximately a factor of two. For example, for a nematic twist structure with  $\psi_0 = \pi/2$ ,  $d = 13 \mu\text{m}$ ,  $\Delta \epsilon / z \sqrt{\epsilon} = 0.17$ , and  $\lambda \equiv 2\pi c / \omega = 0.59 \mu\text{m}$ , van Doorn<sup>28</sup> obtained for the optical threshold a value 1.6 times larger than the critical deformation field. A like value follows from (7.4). The results of other experimental studies, in which the ratio  $E_0 / E_{cr}$  was determined (see, e.g., the detailed measurements of Ref. 7), agree with formula (7.4) with ~10% accuracy. This formula predicts a logarithmic dependence of the ratio  $E_0 / E_{cr}$  on the thickness of the nematic twist structure. In the case of a cholesteric twist structure ( $hd \sim \pi$ ), the dependence of the optical threshold on the layer thickness is substantially differ-

ent [see formulas (7.3) and (7.5)]; a similar dependence was observed in experiments on polarization cutoff of light.<sup>30</sup> We note also that according to (7.4) and (7.5) the optical threshold increases with increasing frequency of the light, since  $\bar{q}_0 \propto \omega \Delta \epsilon(\omega)$ ; this was observed in experiment.<sup>7</sup> Thus, formulas (7.4) and (7.5) are in good agreement with the previously obtained experimental results.<sup>7, 14, 15, 28-31</sup>

The value of the twist angle  $\psi_0$  has little effect on the optical threshold, since the planar structure consists of two symmetrical halves ( $z < d/2$  and  $z > d/2$ ), in each of which the conversion of the light waves takes place independently, and the azimuthal rotation of the director, as noted above, is realized at the center of the layer ( $z \approx d/2$ ). Nonetheless, the result of the combined conversion is strongly affected by the twist angle because of the geometrical-optics interference of the waves.<sup>5</sup> Calculating the "Faraday" integral

$$(\omega/c) \int (\bar{n}_1 - \bar{n}_2) dz$$

[see (2.2)] between the regions of the interaction  $\bar{q} \sim 1$  at  $z < d/2$  and  $\bar{q} \sim 1$  at  $z > d/2$ , we find that the maximum conversion coefficient in the layer is

$$Q_{max} = \sin^2 \psi_0$$

(By definition the conversion coefficient  $Q$  is the ratio of the intensity of the helical wave of one type, emerging from the layer, to the intensity of the wave of the other type incident on the layer.) It is therefore convenient to use in the experiments  $\psi_0 = \pi/2$ , in which case  $Q_{max} = 1$ .

We obtain the width of the optical threshold  $\Delta E_0$  by stipulating that the quantity  $\bar{q}_{min} = \bar{q}(z = d/2)$  change by  $e$  times:

$$\frac{\Delta E_0}{E_0} = \frac{E_{cr} [1 - E_{cr}^2 h^2 d^2 / E_0^2 (\pi^2 - \psi_0^2 + h^2 d^2)]^{1/4}}{E_0 (\pi^2 - \psi_0^2 + h^2 d^2)^{1/4}} \ll \frac{1}{\pi} \frac{E_{cr}}{E_0}. \quad (7.6)$$

It is precisely to this accuracy that formulas (7.4) and (7.5) are valid. The fact that the relative width of the threshold is essentially independent of the twist-structure parameters is due to the exponential character of the deformation [see (7.2)]. If the coupling of the molecules to the walls is not rigid, then the width of the threshold can be less, since the plot of  $\Delta \bar{q}$  against the applied field  $E$  is in this case steeper (see Ref. 36).

## §8. GRANDJEAN TEXTURE IN A LONGITUDINAL FIELD

In a cholesteric twist structure (Grandjean texture), containing several pitches of the free helix ( $h \gg \pi/d$ ), the value of the twist angle  $\psi_0$  does not affect the value of the threshold fields. According to (6.2), the critical deformation field is

$$E_{cr} = (4\pi K_3 h^2 / \Delta \epsilon_0)^{1/4}. \quad (8.1)$$

At  $E > E_{cr}$ , the director in the central part of the layer is inclined to the helix axis  $z$  and, starting with certain values of the field  $E$ , the quantity  $\bar{q}$  (6.9) becomes small compared with unity. This, however, is insufficient for polarization cutoff of the light. In addition to the condition (3.4)  $|\Delta \bar{q}| \sim 1$ , which is satisfied at  $\bar{q}_0 > 1$

[see (6.7)], for cutoff of the light it is necessary that the helix be sufficiently inhomogeneous, i. e., that the rate  $d\tilde{q}/d\psi$  of the change of  $\tilde{q}$  when the director is rotated be large. The last requirement determines the interaction parameter (3.5), which in this case is written in the form<sup>7)</sup> [see (6.10)]

$$G \approx 2\sqrt{2}h(\Delta\epsilon_0 E^2/4\pi K - h^2)^{-1/2}. \quad (8.2)$$

The optical threshold  $E_0$  is obtained from the condition  $G = 1$

$$E_0 \approx 3E_{cr}. \quad (8.3)$$

Obviously, the optical threshold is not sharply pronounced, inasmuch as to decrease the interaction parameter (8.2) by one-half it is necessary to change the field  $E$  by a value of the order of the critical field  $E_{cr}$ . Since  $\omega$  does not enter in the formula (8.2) for the interaction parameter, it is clear that the polarization cutoff of the light sets in immediately in the entire optical band. However, since the Grandjean texture, just as the nematic twist structure, consists of two symmetrical parts, the position of the regions of interaction in which depends on the frequency [ $\tilde{q} = \tilde{q}_0 \sin^2 \alpha \approx 1$ , where  $\tilde{q}_0 \propto \omega \Delta\epsilon(\omega)$ ], the result of the combined conversion also depends on the frequency of the light as a result of the geometrical-optics interference.

## §9. RECIPROCAL TWIST EFFECT

Simultaneous existence of a second-order phase transition and polarization cutoff of light is possible not only in a planar structure but also in a homotropic cholesteric structure, whose boundary walls are so treated<sup>32</sup> that the director  $L$  is perpendicular to them and, consequently, it is directed along the  $z$  axis ( $\alpha = 0$ ) at  $z = 0$  and  $z = d$ . For a cholesteric with  $\Delta\epsilon_0 > 0$  and  $h > \pi K_3/dK_2$ , it can be shown with the aid of relations (2.3) and (3.5) that the critical deformation field coincides approximately with the optical-threshold field. However, owing to the texture transition that reorients the helix axes perpendicular to the field, the case  $\Delta\epsilon_0 > 0$  is of no practical interest (Ref. 2, §§ 6.5, 6.6). We are interested therefore in the present section only in a homotropic structure with a cholesteric having a negative static dielectric anisotropy ( $\Delta\epsilon_0 < 0$ ). Using the de Gennes method (Ref. 1, § 3.2.3) and expression (6.1) for the free energy, we obtain the relation

$$\left| \frac{\Delta\epsilon_0 E_{cr}^2}{4\pi K_3} \right| = \frac{\pi^2}{d^2} - \frac{K_2^2 h^2}{K_3^2} \quad (9.1)$$

which is satisfied by the critical field for the onset of the cholesteric helix. The second-order phase transition that leads to formation of the helix in longitudinal fields  $E > E_{cr}$ , and called the inverse twist effect, is due to reorientation, along the field, of the molecular dipole moments, which for a medium with negative  $\Delta\epsilon_0$  are more or less perpendicular to the long axis of the molecules. [The imaginary value of  $E_{cr}$  (9.1) means that the helix exists even in the absence of a field.]

By virtue of the homotropic boundary conditions, the helix is always free and consequently uniform:

$$d\psi/dz = h; \quad h_0 = 0; \quad \psi_0 = 0 \quad (9.2)$$

[see (6.4)]. The inhomogeneity of the helix is connected with the change of the angle  $\alpha$  between the director  $L$  and the axis of the helix  $z$ , which is described by the equation

$$\left( \frac{d\alpha}{dz} \right)^2 = \left( \frac{|\Delta\epsilon_0| E^2}{4\pi K} + h^2 \right) (\sin^2 \alpha_0 - \sin^2 \alpha) \quad (9.3)$$

[in the approximation (6.5)]. In contrast to (6.3), here  $\alpha_0 = \alpha(d/2)$  is the maximum value of the angle  $\alpha$  at the center of the layer. According to (9.3),  $\alpha_0$  is uniquely connected with the applied field  $E$ :

$$d(|\Delta\epsilon_0| E^2/4\pi K + h^2)^{1/2} = 2F(\sin \alpha_0) \quad (9.4)$$

( $F$  is a complete elliptic integral of the first kind).

For abrupt polarization cutoff of the light, the following conditions must be satisfied:

$$\tilde{q}_0 = \frac{\omega \Delta\epsilon}{4c\sqrt{\epsilon} h} \gg \frac{\pi}{hd} > 1 + \frac{1}{4\tilde{q}_0}. \quad (9.5)$$

The first inequality means that the free cholesteric helix spans many periods of the spatial beats between the helical waves. Otherwise there can be no restructuring of the polarization of the light in the considered smoothly inhomogeneous liquid-crystal structure<sup>17</sup>: if a circularly polarized light wave is incident on the layer (this is precisely how helical waves are polarized at the walls of a homotropic structure), then the wave will leave the layer with practically no change of the polarization.

The second inequality in (9.5), which limits the thickness of the structure to a half-pitch of the helix, makes it possible to obtain in the center of the layer the value of

$$\tilde{q}_{max} = \tilde{q}_0 \sin^2 \alpha_0 \sim 1 \quad (9.6)$$

even at angles  $\alpha_0 \ll 1$ , when according to (9.4)

$$\sin^2 \alpha_0 = \frac{2d^2}{\pi^2} \left( \frac{|\Delta\epsilon_0| E^2}{4\pi K} + h^2 - \frac{\pi^2}{d^2} \right). \quad (9.7)$$

On the other hand if  $\tilde{q}_{max} \gg 1$  even at  $E = 0$ , the interaction parameter in the homotropic structure

$$G = 2\sqrt{2}h \left[ \tilde{q}_{max} \left( \frac{|\Delta\epsilon_0| E^2}{4\pi K} + h^2 \right) \right]^{-1/2} \quad (9.8)$$

is always much less than unity. The latter means that the linear interaction of the helical waves is strong at all values of the field  $E$ . Therefore, just as in the case of very weak interaction, the polarization cutoff of the light cannot be realized by a change of the field  $E$ . (We recall that the threshold effect of the polarization cutoff of the light constitutes a change from geometric-optics propagation of helical waves in the liquid-crystal layer to a propagation such that effective conversion of one helical wave into another takes place.)

It follows from the indicated conditions, at first glance, that the optical threshold corresponds to the field  $E$  at which  $\tilde{q}_{max} \approx 1$ , i. e.,  $\sin^2 \alpha_0 \approx q_0^{-1}$ , see (9.6) and (6.7). Actually, in this case the drop  $|\Delta\tilde{q}|$  (3.4) becomes of the order of unity, because  $\alpha = 0$  at the walls of a homotropic structure, and consequently  $\tilde{q} = 0$ . At the same time, the interaction parameter (3.5)  $G \leq 1$ , inasmuch as at  $\sin^2 \alpha_0 \approx \tilde{q}_0^{-1} \ll 1$  we have, according to

(9.7) and (9.8),

$$|\Delta\epsilon_0|E^2/4\pi K+h^2\approx\pi^2/d^2; G\approx 2\sqrt{2}hd/\pi.$$

A more careful examination however, shows that the above reasoning is valid only at  $hd\sim 1$ . The point is that a homotropic structure contains two symmetrical interaction sections ( $\tilde{q}\sim 1$  at  $z < d/2$  and  $\tilde{q}\sim 1$  at  $z > d/2$ ), and the light cutoff that occurs in the first section can be cancelled by the conversion of the helical waves on the second section.<sup>5</sup> In the general case the cancellation becomes impossible and polarization cutoff of the light appears if the helical waves acquire in the interval between the interaction sections a phase difference of the order of  $\pi/2$ . Using formulas (2.2) and Eq. (9.3), we see that the real optical threshold corresponds to the field  $E_0$  at which  $\tilde{q}_{\max}\approx\pi/hd$ , i.e.,  $\sin^2\alpha_0\approx\pi/\tilde{q}_0hd$ . From this and from (9.7) we obtain the connection between the optical threshold and the critical field of the phase transitions (9.1):

$$E_c^2=E_{cr}^2[1+\pi/2\tilde{q}_0hd(1-h^2d^2/\pi^2)]. \quad (9.9)$$

Obviously, even at  $hd/\pi < 2^{-1/2}$  the relative difference between the fields  $E_0$  and  $E_{cr}$  reaches an extremely small value of the order of  $\pi/\tilde{q}_0hd \ll 1$  [see (9.5)]. It is easy to show that the same quantity determines also the limiting width of the optical threshold. We note also that the polarization cutoff of the light sets in first for blue light and only later for red light, inasmuch as  $\pi/\tilde{q}_0hd \propto 1/\omega\Delta\epsilon(\omega)$ .

We note that in a nematic homotropic structure it is also possible to have a phase transition as well as the above-described effect of polarization cutoff of the light (the inverse twist effect), if "mixed" boundary conditions are produced on both walls of the liquid-crystal structure.<sup>37,38</sup>

## § 10. CHOLESTERIC-NEMATIC TRANSITION

In contrast to the structural phase transitions considered above, the cholesteric-nematic phase transition takes place in a transverse field  $E$  (at  $\Delta\epsilon_0 > 0$ ). For a free helix, the critical field of the transition is well known:

$$E_{cr}=(4\pi K_2\Delta\epsilon_0)^{1/2}\pi^2/g_0$$

(see, e.g., Ref. 1, § 6.2.2 and Ref. 2, §§ 6.5, 6.6). For a non-free cholesteric helix produced in a layer with planar boundary conditions, the process of the field untwisting takes place jumpwise,<sup>20</sup> inasmuch as the number of domain walls decreases in discrete fashion with increasing field  $E$ . As shown in § 5, there is no conversion of helical waves when light passes through one or several domain walls (5.3). Therefore, if the directions of the easy-orientation axes at the boundary walls are parallel (see Fig. 2), then polarization cutoff of the light is impossible. However, threshold polarization cutoff of the light will take place if the directions of the easy orientation axes of the walls are orthogonal (crossed planar structure, see Fig. 4). In this case, in fields close to or larger than critical, the director rotates quite sharply through an angle  $\pi/2$  in accordance with the law (5.3) at one of the boundary walls whose easy-orientation axis is orthogonal to the field (half of

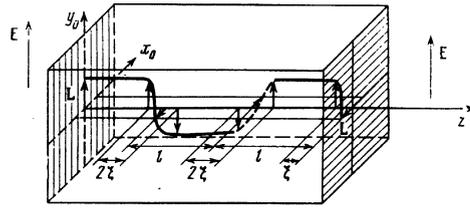


FIG. 4. Strongly untwisted cholesteric helix in a transverse field  $E$  (the directions of the easy-orientation axis on the walls are orthogonal).

domain wall). Such a rotation is typical of any liquid crystal, both nematic and cholesteric, and does not depend on the pitch of the free helix  $g_0=2\pi/h$  (Ref. 1, § 3.2.2). An exact solution of Eqs. (2.8) and (2.10), describing the propagation of light from the region when  $L \parallel E$  into the region where  $L \perp E$ , for high-frequency electric-field components orthogonal to the  $z$  axis, can be written in the form

$$E_{\perp}=\exp\left[-\frac{i\omega}{2c}\int_{z_{in}}^z(\tilde{n}_1+\tilde{n}_2)dz\right]\left\{C_1\left(\operatorname{th}\frac{z}{\xi}-2iq_x\right)\exp\left[\frac{iq_x}{\xi}(z-z_{in})\right]+ \frac{C_2}{\operatorname{ch}(z/\xi)}\exp\left[-\frac{iq_x}{\xi}(z-z_{in})\right]\right\}, \quad (10.1)$$

$$E_{\parallel}=\exp\left[-\frac{i\omega}{2c}\int_{z_{in}}^z(\tilde{n}_1+\tilde{n}_2)dz\right]\left\{-\frac{C_1}{\operatorname{ch}(z/\xi)}\exp\left[\frac{iq_x}{\xi}(z-z_{in})\right]+ C_2\left(\operatorname{th}\frac{z}{\xi}+2iq_x\right)\exp\left[-\frac{iq_x}{\xi}(z-z_{in})\right]\right\}.$$

Here the constants  $C_1$  and  $C_2$  are determined by the boundary conditions

$$C_{1,2}=\frac{E_{\perp,1}^{in}\operatorname{ch}(z_{in}/\xi)+E_{\perp,2}^{in}\operatorname{ch}^2(z_{in}/\xi)[\pm\operatorname{th}(z_{in}/\xi)+2iq_x]}{1+\operatorname{ch}^2(z_{in}/\xi)[\operatorname{th}^2(z_{in}/\xi)+4q_x^2]}.$$

$E_{\perp}$  and  $E_{\parallel}$  are the field components orthogonal and parallel to the director  $L$ , respectively;  $E_{\perp}^{in}$  and  $E_{\parallel}^{in}$  are their values in the region  $z=z_{in}$ , in which the director  $L$  is parallel to the external field  $E$  [more accurately, the vectors  $L$  and  $E$  make a small angle  $\psi(z_{in})$ ]. From this it is easy to find the coefficient of transformation of one helical wave into another:

$$Q=\frac{[(1+q_x^2)^{1/2}-q_x]^2}{2(1+4q_x^2)(1+q_x^2)^{1/2}}, \quad q_x=\tilde{q}_0h\xi=\frac{\omega\Delta\epsilon\xi}{2c(\sqrt{\epsilon_{\parallel}}+\sqrt{\epsilon_{\perp}})}.$$

Inasmuch as the maximum transformation coefficient (as  $q_x \rightarrow 0$ ) is equal to  $1/2$ , the optical threshold is obtained from the condition  $Q \approx 1/4$ , i.e.,  $q_x \approx \sqrt{2}/8$ :

$$E_0=\left(\frac{8\pi K_2}{\Delta\epsilon_0}\right)^{1/2}\frac{\omega\Delta\epsilon}{c\sqrt{\epsilon}}. \quad (10.2)$$

This value of the threshold field can be obtained directly from the qualitative theory of linear interaction, by equating the interaction parameter (3.5) to unity, inasmuch as  $G \approx 4\sqrt{2}q_x$  according to (5.3). The exact solution proves the correctness of the qualitative approach to the investigation of phenomena connected with linear conversion of light.

Comparing the values of the fields  $E_{cr}$  and  $E_0$ , we see that in the case  $\tilde{q}_0 \geq 1$  when the pitch of the free cholesteric helix  $g_0$  exceeds the period  $4\pi c\sqrt{\epsilon}/\omega\Delta\epsilon$  of the spatial beats between the light waves, the optical threshold greatly exceeds the critical field of the cholesteric-ne-

matic transition:  $E_0 = E_{cr} 8\sqrt{2}\bar{q}_0/\pi$ .

On the other hand if the free-helix pitch is small enough ( $\bar{q}_0 \ll 1$ ), then the conversion of the helical waves and the polarization cutoff of the light set in immediately after the formation of the "domain" wall of the helix, characterized by the inequality  $l \gg \xi \approx g_0/\pi^2$  [see (5.2), (5.3), and Fig. 4]. In this case the polarization cutoff of the light is abrupt and takes place practically simultaneously with the cholesteric-nematic transition. The optical threshold  $E_0$  differs from the critical transition field  $E_{cr}$  by a small amount:

$$E_{cr} - E_0 \approx E_{cr} \frac{8\bar{q}_0}{\pi} \ln\left(\frac{4\pi^2 e}{\bar{q}_0^2}\right) \ll E_{cr}.$$

The difference  $E_{cr} - E_0$  was obtained from the condition (3.4)  $\Delta\bar{q} \approx \bar{q}_{max} \sim 1$ , where  $\bar{q}_{max}$  is the maximum value of  $\bar{q}$  (2.3) reached in the interval between the domain walls. The width of the optical threshold is of the order of  $E_{cr} - E_0$ .

## § 11. CONCLUSION

From the content of the present article it follows that the propagation of the light waves in smoothly inhomogeneous liquid crystals of the cholesteric type is accompanied by linear-interaction effects. These effects lead to transformation of one type of helical waves into another, and such a transformation is connected in principle with the helical structure of the crystals. The phenomenon of linear interaction in liquid-crystal optics uncovers a possibility of producing, on a new basis, devices that control the polarization or the intensity of transmitted radiation. These devices will have other frequency characteristics than those known at present. The change of the degree of inhomogeneity of the cholesteric helix, sufficient for the appearance of linear transformation of light, calls for smaller control voltages than that restructuring of the liquid crystal on which the action of present-day liquid-crystal devices is based. The last circumstance gives grounds for hoping also to increase the operating speed. This is particularly important, since high inertia is the main shortcoming of the existing liquid-crystal devices.

On the other hand, our theoretical analysis of threshold effects of polarization cutoff of light points to new possibilities of polarization optical methods of studying liquid-crystal structures and their phase transitions under the influence of an external field. In this respect, particular interest attaches to the structures considered above, in which an optical threshold is present alongside the critical phase-transition point.

As is clear from the foregoing, an abrupt threshold for polarization cutoff of the light is typical of essentially inhomogeneous spiral liquid-crystal layers, in which the pitch of the "free" cholesteric helix is larger than the scale of the inhomogeneity or the thickness of the layer. The difference between the optical threshold and the critical phase-transition point can differ in this case. It is usually large for the twist effect and for the cholesteric-nematic transition in a weakly twisted helix. At the same time, polarization cutoff of the light takes place practically simultaneously with the phase

transition in the case of the reciprocal twist effect and in the case of field untwisting of a cholesteric helix with a sufficiently small pitch. The exact formulas obtained in this article for the optical thresholds make it possible to connect the structural properties of the liquid crystals with their optical properties and to use the results of measurements of the threshold of the phase transition and of the polarization cutoff of the light to determine the parameters of the liquid crystals.

- <sup>1</sup>The results of a numerical calculation of the propagation of polarized light for concrete models of an inhomogeneous strongly twisted helix with pitch  $g \lesssim \lambda$  are given in Refs. 20. Even in a smoothly inhomogeneous plasma with  $\Lambda \gg \lambda$ , owing to the Bragg reflection, interaction of opposing waves takes place, whereas the conversion of the co-moving helical waves is weak.
- <sup>2</sup>It can be shown that complete conversion of one type of wave into another ( $Q = 1$ ) is possible if the helix is strongly twisted at the start and end of the liquid-crystal cell, and the sign of rotation changes at the point  $\zeta = 0$ . In this case  $|\Delta\eta| = \pi/2$  and the transformation effectiveness  $Q \rightarrow 1$  as  $G \rightarrow 0$ .
- <sup>3</sup>In addition to a linear layer (4.1), it is possible also to have a variant wherein the pitch of the helix varies linearly,  $\zeta(\xi) = g'\xi$  ( $0 < \xi < \alpha$ ,  $g' = \text{const}$ ), so that  $\psi' \propto \zeta^{-1}$  ("hyperbolic layer"). In this case the interaction parameter (2.5) is the quantity  $G = 8\sqrt{2} \pi/g'$  and the conversion coefficient varies like  $G = [1 + \exp(\pi G/2\sqrt{2})]^{-1}$ .
- <sup>4</sup>The angle  $\psi_0$  is determined by the azimuthal orientation of the boundary plates and is equal to the differences between the total twist angle of the twist structure and the twist angle of the free helix over the same length. In particular, for a nematic  $h = 0$ , and the angle  $\psi_0$  is equal to the total twist angle. We assume for the sake of argument that  $0 \leq \psi_0 \leq \pi/2$ ; it is actually impossible to reach a value  $\psi_0 > \pi/2$  because of the relaxation of the twisting by structure defects and because of formation of disclinations.
- <sup>5</sup>We are considering the free energy for the electric field at a fixed voltage on the outer conductors and for a given volume and number of particles of the liquid crystal. Frequently, to prevent charge transport and electrohydrodynamic instability, an alternating electric field of low frequency is used (variation of this frequency makes it possible also to regulate the dielectric anisotropy  $\Delta\epsilon_0$ ). If the external field is magnetic, then the last term in (6.1) must be replaced by  $\Delta\chi H^2/2$ , where  $\Delta\chi$  is the static magnetic anisotropy and  $H$  is the intensity of this field. For a magnetic field, all the formulas that follow are valid subject to this substitution.
- <sup>6</sup>Here and below it is assumed that in the considered range of electric fields  $E$  there are no electrohydrodynamic instabilities. The corresponding conditions are usually realized in numerous experiments with twist cells,<sup>7, 14, 15, 28, -31</sup> in which the polarization cutoff of light is observed at voltages lower than the threshold of indicated instabilities. On the other hand, if the electrohydrodynamic instability sets in before the structural phase transition, then the polarization optical properties of the texture can be substantially different. We note that no such instabilities occur at all in a magnetic field.
- <sup>7</sup>We have used here the inequality  $8\bar{q}_0 \leq \exp(hd)$ , which is actually always satisfied by virtue of the condition  $h \gg \pi/d$ .

<sup>1</sup>P. de Gennes, *The Physics of Liquid Crystals*, Oxford, 1974.

<sup>2</sup>L. M. Blinov, *Élektro- i magnitooptika zhidkikh kristallov* (Electro- and Magneto-optics of Liquid Crystals), Nauka (1978).

<sup>3</sup>V. L. Ginzburg, *Zh. Tekh. Fiz.* 14, 181 (1944).

<sup>4</sup>E. V. Suvorov, *Izv. Vyssh. ucheb. zaved. Radiofizika* 15,

- 1320 (1972). N. S. Bellyustin, *ibid.* **23**, 133 (1980).
- <sup>5</sup>V. V. Kocharovskii and V. V. Kocharovskii, *Fizika plazmy* **6**, 565 (1980) [*Sov. J. Plasma Phys.* **6**, 308 (1980)].
- <sup>6</sup>V. A. Belyakov, V. E. Dmitrienko and V. P. Orlov, *Usp. Fiz. Nauk* **127**, 221 (1979) [*Sov. Phys. Usp.* **21**, 63 (1979)].
- <sup>7</sup>M. F. Grebenkin, V. A. Seliverstov, L. M. Blinov and V. G. Chigrinov, *Kristallografiya* **20**, 984 (1975) [*Sov. Phys. Crystallography* **20**, 604 (1975)].
- <sup>8</sup>Shoichi Matsumoto *et al.* *J. Appl. Phys.* **47**, 3842 (1976).
- <sup>9</sup>V. Freedericksz and V. Zolina, *Trans. Far. Soc.* **29**, 919 (1933).
- <sup>10</sup>V. Freedericksz and V. Tsvetkov, *Phys. Z. Sovjetunion* **6**, 490 (1934).
- <sup>11</sup>E. Guyon, *Amer. J. Phys.* **43**, 877 (1975).
- <sup>12</sup>E. Sackmann, S. Meiboom and L. C. Snyder, *J. Amer. Chem. Soc.* **89**, 5981 (1967).
- <sup>13</sup>J. Wysocki, J. Adams and W. Haas, *Phys. Rev. Lett.* **20**, 1024 (1968). H. Baessler and M. M. Labes, *ibid.* **21**, 1791 (1968).
- <sup>14</sup>C. J. Gerritsma, W. H. de Jen and P. van Zenten, *Phys. Lett.* **36A**, 389 (1971).
- <sup>15</sup>M. Schadt and W. Helfrich, *Appl. Phys. Lett.* **18**, 127 (1971).
- <sup>16</sup>V. V. Zheleznyakov, *Elektromagnitnye volny v kosmicheskoi plazme (Electromagnetic Waves in Cosmic Plasma)*, Nauka (1977), §7.
- <sup>17</sup>V. V. Zheleznyakov, V. V. Kocharovskii and V. I. Kocharovskii, *Zh. Eksp. Teor. Fiz.* **77**, 101 (1979) [*Sov. Phys. JETP* **50**, 51 (1979)].
- <sup>18</sup>E. I. Kats, *Zh. Eksp. Teor. Fiz.* **59**, 1864 (1970) [*Sov. Phys. JETP* **32**, 1004 (1970)].
- <sup>19</sup>V. A. Kizel', Yu. I. Krasilov and V. I. Burkov, *Usp. Fiz. Nauk* **114**, 295 (1974) [*Sov. Phys. Usp.* **17**, 745 (1975)], Yu. V. Denisov, V. A. Kizel', E. P. Sukhenko and V. G. Tishchenko, *Kristallografiya* **21**, 991 (1976); **22**, 339 (1977).
- [*Sov. Phys. Crystallogr.* **21**, 568 (1976); **22**, 192 (1977)].
- <sup>20</sup>R. Dreher, *Sol. St. Comm.* **12**, 519 (1972), L. E. Hajdo and A. C. Eringen, *JOSA* **69**, 1017 (1979).
- <sup>21</sup>D. B. Melrose, *Aust. J. Phys.* **27**, 31 (1974).
- <sup>22</sup>K. G. Budden, *Radio waves in the ionosphere*, Cambridge Univ. Press (1961).
- <sup>23</sup>R. Cano, *Bull. Soc. fr. Miner. Crystallogr.* **90**, 333 (1967).
- <sup>24</sup>Iu. A. Kravtsov, *Dokl. Akad. Nauk SSSR* **183**, 74 (1968) [*Sov. Phys. Dokl.* **13**, 1125 (1969)].
- <sup>25</sup>E. Kamke, *Differenzialgleichungen*, Bd. 1. Chelsea, 1971 (Russ. transl. Nauka, 1976).
- <sup>26</sup>F. Leenhouts and Van der Woude, *J. de Phys. Lett.* **39**, 249 (1978).
- <sup>27</sup>V. E. Dmitrienko and V. A. Belyaev, *Zh. Eksp. Teor. Fiz.* **78**, 1568 (1980) [*Sov. Phys. JETP* **51**, 787 (1980)].
- <sup>28</sup>C. Z. van Doorn, *Phys. Lett.* **42A**, 537 (1973).
- <sup>29</sup>A. Boller *et al.* *Proc. IEEE* **60**, 1002 (1972).
- <sup>30</sup>A. Ashford *et al.* *Electron. Lett.* **9**, 118 (1973).
- <sup>31</sup>E. P. Raynes, *Electron. Lett.* **9**, 101 (1973), **10**, 131 (1974).
- <sup>32</sup>F. J. Kahn, G. N. Taylor, and H. Schonhorn, *Proc. IEEE* **61**, 823 (1973).
- <sup>33</sup>C. H. Gooch and H. A. Tarry, *Electr. Lett.* **10**, 2 (1974), *J. Phys.* **8D**, 1575 (1975).
- <sup>34</sup>C. Mauguin, *Bull. Soc. Franc. Min.* **34**, 3 (71) (1911), *Phys. Z.* **12**, 1011 (1911).
- <sup>35</sup>J. Nehring, *Phys. Lett.* **72A**, 446 (1979).
- <sup>36</sup>J. Nehring, A. R. Kmetz and T. J. Scheffer, *J. Appl. Phys.* **47**, 850 (1976).
- <sup>37</sup>L. M. Blinov, *Usp. Fiz. Nauk* **114**, 67 (1974) [*Sov. Phys. Usp.* **17**, 658 (1975)].
- <sup>38</sup>F. Ogawa, C. Tani and F. Saito, *Electron. Lett.* **12**, 70 (1976).

Translated by J. G. Adashko

## Complete population inversion in a multilevel quantum system on adiabatic application of an external resonance field

M. V. Kuz'min and V. N. Sazonov

*P. N. Lebedev Physics Institute, Academy of Sciences of the USSR, Moscow*  
(Submitted 28 April 1980)  
*Zh. Eksp. Teor. Fiz.* **79**, 1759-1768 (November 1980)

When an external resonance field is applied sufficiently slowly, a multilevel quantum system can be transferred from the initial ground state to any other higher level and the probability of this process is close to unity. However, this excitation mechanism is impossible in the case of a two-level system. The interval of external field frequencies in which a system can be excited does not increase when the field intensity is increased and, in principle, can be made as small as we please, which ensures a high excitation selectivity. This inversion mechanism acts at quite realistic values of the external field intensity and of the rate of its rise.

PACS numbers: 42.55.Bi

### 1. INTRODUCTION

Most of the investigations of the resonance action of radiation on quantum systems have been carried out using a two-level model for which it is quite easy to obtain a clear analytic solution valid in the resonance approximation or in the approximation of a rotating wave.<sup>1-3</sup> Recently, even books have been devoted entirely to the subject of the two-level model (see, for example, Ref. 3). However, one can justifiably use

the two-level model only as long as it describes correctly all the qualitative features of real systems and the model has to be refined in quantitative features of real systems and the model has to be refined in quantitative calculations. We shall show that multilevel systems can exhibit a certain qualitatively different effect which does not occur in two-level systems. This effect is as follows: when an external resonance field is applied sufficiently slowly, a system initially in the ground quantum state can be transferred to a higher level with