

Optics of cholesteric liquid crystals

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A review is presented of the theory of optical properties of cholesteric liquid crystals (CLC). The diffraction nature of the unusual optical properties of CLC is exhibited in the simplest and most easily visualizable kinematic approximation of diffraction theory. A quantitative description of the optical properties is given based on the exact solution of Maxwell equations for light propagated along the optical axis of CLC and within the framework of the dynamic diffraction theory for an arbitrary direction of propagation. Considerable attention is devoted to the manifestation of the connection between the structural and optical properties of the CLC observed when light is propagated at an angle to the optical axis of the CLC and when the CLC structure is distorted by an external field. Investigations of the optical properties of absorbing CLC reveal that suppression of light absorption in the selective reflection band is analogous in its nature to the well-known Borrmann effect in x-ray diffraction. Different approaches to the optics of imperfect CLC which frequently most nearly correspond to the conditions of actual experiments are described. A theory of the optical properties of chiral smectic crystals is presented. This theory, on the whole, is analogous to that for CLC, but it predicts a number of qualitative differences in the optics of chiral smectic crystals. The theory and special features of the Vavilov-Cherenkov radiation in CLC is briefly presented. A comparison is made of the theoretical results with the experimental data and the most promising directions for theoretical and experimental research are indicated.

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1. INTRODUCTION

The recent steady interest in liquid crystals is due to the very interesting physics of the liquid-crystalline state,¹⁻⁹ to their role in biological systems,^{10,11} and last but not least, to the broad potentialities of their varied applications. Among these applications, primarily the unusual optical properties of liquid crystals are employed, and in particular, the possibility of controlling these properties by small perturbations (electric, magnetic, mechanical, etc.).¹²⁻¹⁴ Three varieties of liquid crystals are known: nematic (NLC), smectic (SLC) and cholesteric (CLC). Among these, the cholesteric liquid crystals possess the most remarkable and varied optical properties.^{5,15-20}

This review is concerned with the optics of cholesteric and in part the chiral phase of smectic, liquid crystals. Up to now a large number of experimental and theoretical studies has been published on this topic, and advances have been able to be made not only in qualitative understanding of the unusual optical properties of CLCs, but also in describing them quantitatively. At least as regards perfect specimens of CLCs, the problem in theoretical description of the optical properties is no longer that of revealing the physical nature of the observed phenomena, but of choosing which of the developed theoretical approaches best corresponds to the situation under study. Nevertheless, in spite of marked progress, it is rather complicated to get a global view of the subject (especially for those beginning to interest themselves in this problem). The reason for this is that the optics of CLCs is presented too sketchily in the monographs and reviews of general type,¹⁻²⁰ while the numerous original articles are devoted to special problems and often duplicate one another, which also does not always foster clarity in gaining acquaintance with the topic. Yet undoubtedly, the problems of the optics of CLCs merit an independent presentation, since a unique situation is realized among the CLCs for wavelengths of the optical range in which light is diffracted by the complicated spatial structure of the CLCs. In essence the special and unusual character of the optical properties of CLCs proves to involve diffractive light scattering.

This article aims to present from a unitary outlook the state of the theory of the optical properties of cholesteric liquid crystals and to compare its conclusions with the results of experimental studies. It seems to us that an acquaintance with the achievements and problems of the optics of CLCs can be useful not only to specialists on liquid crystals, but also to a broader set of persons interested in problems of coherent interaction of various types of radiation with crystals.

CLCs possess remarkable optical properties.^{5,15-20} They selectively reflect light of a definite polarization and definite wavelength. Thus, for example, when a ray is incident on a CLC along the optic axis, light of one circular polarization in a relatively narrow range of wavelengths is reflected from the crystal, while light of the other (orthogonal) circular polariza-

tion penetrates into the crystal without being reflected. The color of a CLC depends on the angle at which the crystal is viewed. In thin layers the observed rotation of the plane of polarization of light is as much as several thousand degrees per millimeter. This cannot be explained by the natural optical activity of the molecules. Imposing external fields (electric, magnetic, etc.) or changing the temperature, alters the wavelength of light at which the described anomalous optical properties are manifested.

The nature of the optical peculiarities of a CLC proves to involve its structure, which for monocrystalline layers is that shown in Fig. 1. The long axes of the molecules forming the CLC, which are depicted in Fig. 1 by oval dots, are oriented in the same way in any plane perpendicular to the optic axis (*z* axis), but without long-range order in the arrangement of the centers of gravity of the molecules. The orientation of the axes of the molecules as a function of the coordinates varies according to the law

$$\varphi(z) = \frac{2\pi z}{p} \tag{1.1}$$

Here $\varphi(z)$ is the rotation angle of the axis of the molecule in the plane. The quantity p is called the period or pitch of the cholesteric helix. In a distance p along the *z* axis, the axes of the molecules rotate by an angle of 2π . A characteristic value of p for most CLCs amounts to several hundred or thousand Ångströms, though CLCs are found with larger values of p .

The pattern depicted above of the structure of CLCs is idealized. In particular, the orientations of molecules at a fixed value of *z* can differ slightly from one another, while Eq. (1.1) describes the *z*-dependence of the mean orientation of the molecules. The direction of the mean orientation of the molecules is commonly described by the unit vector **n**, which is called the director. The degree of order of the molecules is characterized by the order parameter^{3,5}

$$S = \frac{1}{2} (3 \langle \cos^2 \theta \rangle - 1) \tag{1.2}$$

Here $\langle \cos^2 \theta \rangle$ is the mean square of the cosine of the deviation angle of the long axes of the molecules from the direction **n** of the director. Thus Eq. (1.1) describes the spatial variation of the orientation of the director. When $S \neq 1$, the latter coincides only on the

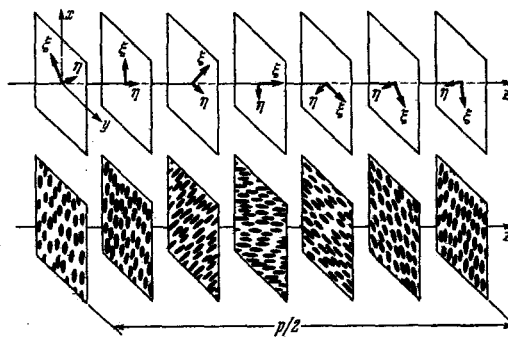


FIG. 1. Schematic drawing of the structure of a cholesteric liquid crystal (below) and the corresponding dependence on the coordinates of the orientation of the principal axes of the dielectric permittivity tensor (above).³

average with the orientation of the axes of the molecules.

A cholesteric phase (or, as is commonly said, a cholesteric mesophase) can exist for organic compounds that have elongated molecules without mirror symmetry (chiral). Typical representatives of these compounds are the derivatives of cholesterol. Thence arose the name of cholesteric crystals. A cholesteric phase usually exists over a certain temperature range directly from the melting point of the ordinary crystalline phase up to a higher temperature that corresponds to the cholesteric liquid transition. Usually the pitch of the cholesteric helix declines with increasing temperature, though the opposite trend in the pitch can occur in some cases. A certain particular compound can also have a smectic and a cholesteric phase.^{3,5} In these cases the smectic phase precedes the cholesteric. That is, a smectic liquid crystal first arises on melting of the crystal, and then a phase transition to a CLC occurs with increasing temperature.

The amount of twist of a cholesteric phase is very small on a molecular scale ($a/p \sim 10^{-2} - 10^{-3}$, where a is the characteristic dimension of the molecules). This stems from the smallness of the deviation of the symmetry properties of the molecules from mirror symmetry. A detailed presentation of the physics of a cholesteric mesophase lies outside the scope of this article, and it can be found, for example, in some recently published books.^{5,13}

The nature of the anomalous optical properties of CLCs (such as the selective reflection of light of a certain polarization and wavelength, the unique high rotation of the plane of polarization of light in thin layers, the dependence of the color of the specimen on the temperature and also on the angle of observation, etc.) has been understood in general terms for a rather long time, already in the thirties, following the work of Oseen.^{21,22} The physical cause of the stated "anomalies" in the optical properties of CLCs is the diffraction of light by their spatially periodic structure (cholesteric helix), whose period usually lies in the region of the optical wavelength range. This explains naturally the variation with temperature (or with imposition of an external field) of the wavelength range in which the optical anomalies are observed. Actually, the pitch p of the cholesteric helix is altered by the stated agents, and this leads to the observable consequences. However, in spite of the fact that the diffractive nature of the optical anomalies of CLCs has occasioned no doubt, a number of important quantitative characteristics and qualitative effects that are manifested in the optics of CLCs have only recently found an adequate theoretical description.

The most natural way to describe the optical properties of CLCs consists of solving the Maxwell equations (with an appropriate assignment of boundary conditions), in which the dielectric properties of the CLCs are described by the coordinate-dependent dielectric-constant tensor $\hat{\epsilon}(\mathbf{r})$.⁵ The coordinate-dependence of the tensor $\hat{\epsilon}$ consists in the point-to-point variation of the

orientation of the principal axes of the tensor, the local direction of which is determined by the orientation of the molecules of the CLC at the given point.

For a perfect cholesteric structure $\hat{\epsilon}(\mathbf{r})$ has the form

$$\hat{\epsilon}(\mathbf{r}) = \begin{pmatrix} \bar{\epsilon} + \bar{\delta} \cos \tau z & \pm \bar{\delta} \sin \tau z & 0 \\ \pm \bar{\delta} \sin \tau z & \bar{\epsilon} - \bar{\delta} \cos \tau z & 0 \\ 0 & 0 & \epsilon_3 \end{pmatrix}. \quad (1.3)$$

Here the z axis lies along the optic axis, we have $\tau = 4\pi/p$, p is the pitch of the cholesteric helix, ϵ_1 and $\epsilon_2 = \epsilon_3$ are the principal values of the dielectric-constant tensor, $\bar{\epsilon} = (\epsilon_1 + \epsilon_2)/2$, and $\bar{\delta} = (\epsilon_1 - \epsilon_2)/(\epsilon_1 + \epsilon_2)$. As we see from (1.3), the period of variation of the dielectric properties of the CLC is half the pitch. The two signs in (1.3) correspond to the two geometric possibilities: plus to a right-hand and minus to a left-hand cholesteric helix.

The difficulty in solving the problem formulated above is determined by the complicated form of the tensor $\hat{\epsilon}$. However, on the whole we should state that currently a complete physical understanding of the unusual optical properties of CLCs has been attained. Yet the choice of a method of optical theoretical description of an experiment is determined by its specific conditions and required accuracy. In line with the fact that the anomalies in the optical properties of CLCs are directly manifested in Vavilov-Čerenkov radiation, this review also presents the theory of coherent emission by fast charged particles in CLCs.

2. KINEMATICAL TREATMENT

Many of the above-listed anomalies in the optical properties of CLCs follow from an elementary, so-called kinematical, treatment of the diffraction of light by the periodic structure of a CLC. The corresponding approach is applicable in the case of low intensity of the scattered beam as compared with the incident beam, and has been well developed and widely applied in describing scattering by crystals of x-rays and of thermal neutrons,^{23,24} while its results admit a clear physical interpretation. Hence we shall start the description of the optical properties of CLCs with the kinematical approximation.

a) The scattering cross-section

For the sake of concreteness, we shall treat the transmission and reflection of light for the case of a so-called plane structure, i.e., a monocrystalline specimen of a CLC in the form of a plane-parallel plate with its optic axis (z axis) perpendicular to the surface (Fig. 2). In order to simplify the treatment, we shall not take into account here reflection of light at the boundaries that is due simply to the differing values of the refractive index inside and outside the specimen.

In the kinematical approximation, the light-scattering cross-section of a CLC is described by the following expression (see, e.g., Refs. 5 and 25):

$$\frac{d\sigma(\mathbf{k}_0, \mathbf{e}_0; \mathbf{k}_1, \mathbf{e}_1)}{d\Omega_{\mathbf{k}_1}} = \left(\frac{\omega^2}{4\pi c^2} \right)^2 \times \left| \int (e_1^* \hat{\epsilon} e_0) \exp[i(\mathbf{k}_0 - \mathbf{k}_1) \cdot \mathbf{r}] d\mathbf{r} \right|^2. \quad (2.1)$$

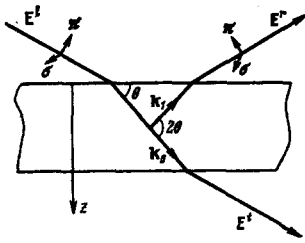


FIG. 2. Geometry of scattering by a plane structure of a CLC. The optic axis of the CLC (z axis) is perpendicular to the surface of the specimen.

Here k_0 , k_1 , e_0 , and e_1 are the wave vectors and polarization vectors of the incident and scattered waves, while we integrate over the volume of the specimen. Let us employ a Fourier expansion for $\hat{\epsilon}(\mathbf{r})$ of (1.3):

$$\hat{\epsilon}(\mathbf{r}) = \sum_{s=-\infty}^{\infty} \hat{\epsilon}_s \exp(i s \tau \mathbf{r}). \quad (2.2)$$

Here we have

$$\hat{\epsilon}_0 = \begin{pmatrix} \bar{\epsilon} & 0 & 0 \\ 0 & \bar{\epsilon} & 0 \\ 0 & 0 & \epsilon_3 \end{pmatrix}, \quad \hat{\epsilon}_1 = \hat{\epsilon}_{-1} = \frac{\bar{\epsilon} \delta}{2} \begin{pmatrix} 1 & \mp i & 0 \\ \mp i & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \hat{\epsilon}_s = 0 \text{ when } |s| > 1.$$

Also, τ is the reciprocal-lattice vector of the CLC ($\tau = (z/|z|)4\pi/p$). From (2.1) and (2.2) we obtain

$$\frac{d\sigma(k_0, e_0; k_1, e_1)}{d\Omega_{k_1}} = \left(\frac{\omega^2}{4\pi c^2} \right)^2 |e_1^* \hat{\epsilon}_s e_0|^2 \left| \int \exp[i(k_0 - k_1 + s\tau) \cdot \mathbf{r}] d\mathbf{r} \right|^2. \quad (2.3)$$

In the limit of infinite specimen dimension, the integral in (2.3) proves to be proportional to the delta function $\delta(k_0 - k_1 + s\tau)$, and the scattering cross-section acquires the form that is well known from the theory of x-ray and neutron scattering.^{23,24} In particular, the scattering directions are determined by the purely kinematical relationship known as the Bragg condition, which tells nothing about the intensity and polarization properties of the scattering:

$$k_1 - k_0 = s\tau, \quad |k_1| = |k_0| \quad \text{or} \quad \sin \theta = \frac{s\lambda}{p}. \quad (2.4)$$

Here 2θ is the scattering angle (see Fig. 2), and $\lambda = 2\pi c/\omega\sqrt{\bar{\epsilon}}$ is the wavelength of the light. We note, strictly speaking, that we should employ in Eqs. (2.1)–(2.4) the values of the wave vectors and polarization vectors inside the specimen. In particular, we have $k_1^2 = k_0^2 = (\omega/c)^2 \bar{\epsilon}$, while the directions of the wave vectors in the specimen and outside it are related by the ordinary laws of refraction.

However, the dependence of the intensity and polarization characteristics of the scattering on the structural details of the specimen is governed by the structure factor

$$F(k_0, e_0; k_1, e_1) = e_1^* \hat{\epsilon}_s e_0. \quad (2.5)$$

The quantity F is an analog of the x-ray structure amplitude²³ and in a CLC it differs from zero only for the first order of diffractive reflection ($s = 1$).

Equation (2.4) explains the dependence of the color of a CLC on the angle of observation and also the change in color of the specimen upon a temperature change of the CLC. The different angles θ of observation correspond to different wavelengths diffractively

reflected by the crystal, i.e., different colors. In particular, at normal incidence light with the wavelength $\lambda = p$ undergoes diffractive reflection. In Fig. 3 the case of normal incidence corresponds to the situation in which k_0 and k_1 lie in opposite directions. That is, the selective reflection of light at normal incidence is simply a special case of diffractive reflection. The variation with temperature of the color of a CLC is also explained by Eq. (2.4), and it simply corresponds to a change in the wavelength of light diffracted by the CLC caused by the temperature-dependence of the pitch p of the cholesteric helix.

b) Polarization characteristics

Equation (2.3) also explains the dependence on polarization of reflection and transmission of light in a CLC. Let us choose as one of the polarization vectors the unit vector σ in the direction perpendicular to the $k_0 k_1$ plane and write the polarization vectors e_0 and e_1 in the form

$$e_0 = \sigma \cos \alpha_0 + i\pi_0 \sin \alpha_0, \quad e_1 = \sigma \cos \alpha_1 + i\pi_1 \sin \alpha_1. \quad (2.6)$$

Here the vectors σ , π_0 and k_0 (σ , π_1 , and k_1) form a right-handed triplet. Here Eq. (2.5) acquires the form

$$F(k_0, e_0; k_1, e_1) = -\frac{\bar{\epsilon} \delta}{2} (\cos \alpha_1 \pm \sin \alpha_1 \sin \theta) (\cos \alpha_0 \pm \sin \alpha_0 \sin \theta). \quad (2.7)$$

Upon obtaining the extrema of Eq. (2.7) with respect to α_0 , we find that the CLC diffractively scatters elliptically polarized light for which the parameter $\alpha_0 = \alpha d/0$ in (2.6) depends on the angle θ and is given by the expression

$$\alpha_0^d = \pm \arctan(\sin \theta). \quad (2.8)$$

The orthogonal polarization $\alpha_0^d = \mp \arctan(1/\sin \theta)$ does not interact with the CLC. The polarization of the scattered wave also depends on the angle of incidence and corresponds to the parameter $\alpha_1 = \alpha d/1 = \alpha d/0$. That is, it is also determined by Eq. (2.8).

Thus the kinematical approach explains also the selective reflection and transmission of differently polarized waves and their dependence on the angle of incidence. In the special case of normal incidence ($\theta = \pi/2$), circularly polarized light is selectively scattered in a CLC; left circularly polarized light in the case of a left-hand cholesteric helix and right circularly polarized light in the case of a right-hand helix.

The kinematical approximation that we have presented here gives a clear physical picture of the nature of the peculiarities of the optics of CLCs and a number of relationships useful for describing experiments. However, a quantitative description of experiment based

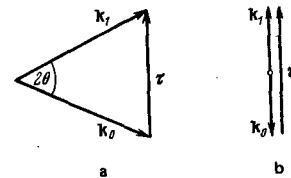


FIG. 3. Illustration of the diffractive nature of the selective scattering of light in a CLC: the Bragg condition with oblique (a) and normal (b) incidence of light on the specimen.

on it can be made only in situations when the dimensions of the regions that scatter light coherently are small, so that we can neglect the attenuation of the light by scattering within a coherently scattering region. This situation is realized either in very thin perfect specimens $[(L\delta/p) \ll 1]$, where L is the thickness of the specimen] or in polycrystalline CLC specimens having small dimensions of the individual crystallites.

At large crystal thicknesses at which multiple Bragg scattering processes begin to play an appreciable role, the kinematical approximation is not suited for quantitative description of experiments nor even for explaining a number of qualitative effects, e.g., the higher orders of diffractive reflection, the rotation of the plane of polarization of light, etc.

Taking single scattering into account does not suffice to explain the cited phenomena, and we need a more rigorous treatment of the interaction of light with the CLC. The following sections will present the results that fall outside the scope of the kinematical approximation.

3. THE CASE OF NORMAL INCIDENCE

As we have noted above, the solutions of the Maxwell equations in a CLC are extremely complicated in the general case. Therefore we shall begin the presentation of results that fall outside the kinematical approximation with the simplest case of propagation of light along the optic axis of a CLC. This case is of great interest, both with respect to the wealth of experimental studies concerned with it, and because an exact solution is known for it. Moreover, the analysis of the solutions conducted for this case proves useful for studying and understanding the solutions in the general case.

a) Eigenwaves

Following the studies of Oseen,^{21,22} De Vries,²⁶ and Kats,²⁷ let us derive the solution of the Maxwell equations in a CLC having the dielectric permittivity of (1.3). For a wave propagating along the optic axis, the equations assume the form

$$\frac{\partial^2 \vec{E}}{\partial z^2} = \hat{\epsilon} \frac{\partial^2 \vec{E}}{\partial t^2}. \quad (3.1)$$

Here the z axis lies along the optic axis, and \vec{E} is the electric field vector in the medium, which in this case is perpendicular to the z axis. We shall seek the field in the crystal in the form of a superposition of two plane waves:

$$\vec{E} = n_+ E_+ \exp \left[i \left(\beta + \frac{\tau}{2} \right) z - i\omega t \right] + n_- E_- \exp \left[i \left(\beta - \frac{\tau}{2} \right) z - i\omega t \right]. \quad (3.2)$$

Here $n_{\pm} = (\sigma \pm i\pi_0)/\sqrt{2}$ are the circular-polarization vectors, and ω is the frequency of the light. That is, we are seeking the solution in the form of Bloch waves, as the periodicity of the CLC requires. We get the following system of equations for the amplitudes E_+ and E_- from (3.1) and (3.2):

$$\begin{aligned} \left[\kappa^2 - \left(\beta + \frac{\tau}{2} \right)^2 \right] E_+ + \kappa^2 \delta E_- &= 0, \\ \kappa^2 \delta E_+ + \left[\kappa^2 - \left(\beta - \frac{\tau}{2} \right)^2 \right] E_- &= 0. \end{aligned} \quad (3.3)$$

Here we have $\kappa^2 = \omega^2 \bar{\epsilon} / c^2$. The system (3.3) has non-zero solutions when its determinant vanishes, i.e.,

$$\left[\kappa^2 - \left(\beta + \frac{\tau}{2} \right)^2 \right] \left[\kappa^2 - \left(\beta - \frac{\tau}{2} \right)^2 \right] - \kappa^4 \delta^2 = 0. \quad (3.4)$$

We get from (3.4) an expression defining β as a function of the frequency of the wave, the period of the helix, and the anisotropy parameter δ :

$$\beta_j = \pm \sqrt{\kappa^2 + \frac{\tau^2}{4} \pm \kappa \sqrt{\tau^2 + \kappa^2 \delta^2}}, \quad j=1, 2, 3, 4. \quad (3.5)$$

Figure 4 shows the numbering of the solutions for β_j in (3.5) and their frequency-dependence. The four solutions (3.5) of Eq. (3.4) define four possible (for a given frequency ω) superpositions of the waves of (3.2) (we shall call them the eigensolutions). Each has its own ratio of the amplitudes E_+ and E_- :

$$\xi_j = \left(\frac{E_-}{E_+} \right)_j = \frac{\kappa^2 \delta}{[\beta_j - (\tau/2)]^2 - \kappa^2}. \quad (3.6)$$

Equations (3.2), (3.5), and (3.6) imply that the two eigensolutions 1 and 4 [which correspond to the plus sign under the radical in (3.5)] correspond to waves that interact weakly with the CLC and which propagate respectively along the z axis and in the opposite direction. The other two eigensolutions 2 and 3, which correspond to the minus sign under the radical, each amount to a superposition of two waves that interact strongly with the CLC and which propagate along the z axis and in the opposite direction. Here the wave propagated along z is represented more strongly in one of the solutions, and that propagated in the negative direction of z in the other. The waves E_+ and E_- in the eigensolutions prove to be circularly polarized in a direction opposite to the screw sense of the cholesteric helix for the "weakly interacting solutions" and in a direction matching that of the helix for the "strongly interacting" solutions. The solutions 2 and 3 of Eq. (3.4) become imaginary near the Bragg frequency $\omega_B = \tau c / 2 \sqrt{\bar{\epsilon}}$, in the frequency range $\omega_B / \sqrt{1+\delta} < \omega < \omega_B / \sqrt{1-\delta}$. Waves of the corresponding circular polarization cannot propagate in the crystal (i.e., there is a "forbidden band"; see Fig. 4). For the solutions in the forbidden band we find that $|E_-/E_+|_{2,3} = 1$. As we shall see below, the selective reflection of light corresponding to Bragg diffractive scattering occurs in this range (see Fig. 5 below).

We stress that, when light propagates along the optic axis, only light having a circular polarization matching the screw sense of the cholesteric helix will undergo diffractive reflection. Here reflection in the higher

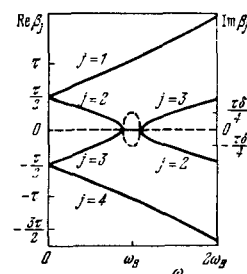


FIG. 4. Frequency dependence of the roots of the dispersion equation when light propagates along the axis of the CLC. Solid curve— $\text{Re } \beta_j$; dotted curve— $\text{Im } \beta_j$.

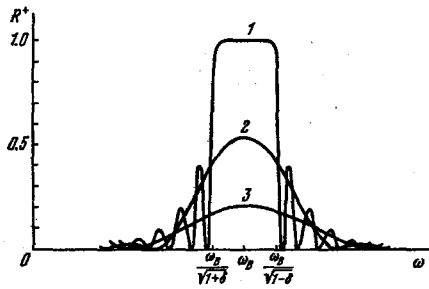


FIG. 5. Reflection coefficient of light in diffraction of circularly-polarized light with normal incidence for different specimen thicknesses. 1— $L\delta/p \gg 1$ (thick specimen), 2— $L\delta/p \sim 1$, 3— $L\delta/p \ll 1$ (thin specimen).

orders that correspond to $k_1 - k_0 = s\tau$ with $s \geq 2$ is absent. The latter statement reflects the fact that the parameter β_j in the obtained exact solution proves to be imaginary in a single frequency range near ω_B .

b) Solution of the boundary problem

In order to find the amplitudes of the wave reflected by the CLC and of that transmitted through the specimen, we must determine which of the four solutions of the system (3.3) are excited by the wave incident on the crystal. This is done by using the boundary conditions that are imposed on both sides of the liquid-crystal plate (see Fig. 2). For the case of normal incidence these conditions consist of the requirement that the electric field \vec{E} and the magnetic field $\vec{H} = (ic/\omega) \text{curl } \vec{E}$ are continuous at the boundaries of the crystal.

Let the wave $\vec{E}^i = (E_+^i \mathbf{n}_+ + E_-^i \mathbf{n}_-) e^{i(\kappa_0 z - \omega t)}$ be incident on the crystal, where κ_0 is the wave vector in the medium surrounding the crystal, and E_+^i and E_-^i are the amplitudes of the right- and left-polarized components in the incident wave. The general solution of Eq. (3.1) for the field in the crystal has the form

$$\vec{E}(\mathbf{r}, t) = e^{-i\omega t} \sum_{j=1}^4 (E_{+j})_j(n_+ e^{i[\beta_j + (\tau/2)]z} + \xi_j n_- e^{i[\beta_j - (\tau/2)]z}). \quad (3.7)$$

We seek the amplitudes of the reflected and transmitted waves in the form

$$\vec{E}^r = (E_+^r \mathbf{n}_+ + E_-^r \mathbf{n}_-) e^{-i(\kappa_0 z + \omega t)}, \quad \vec{E}^t = (E_+^t \mathbf{n}_+ + E_-^t \mathbf{n}_-) e^{i(\kappa_0 z - \omega t)}. \quad (3.8)$$

Here E_+^r and E_-^r (E_+^t and E_-^t) are the amplitudes of the dextro- (and levo-) polarized components. Employing the continuity of \vec{E} and \vec{H} at the boundaries of the crystal, we obtain the following expression for E_+^r :

$$E_+^r = -D^{-1} \times \begin{vmatrix} a_{11} & a_{12} & a_{13} & a_{14} & E_+^i \\ a_{21} & a_{22} & a_{23} & a_{24} & E_-^i \\ a_{31} & a_{32} & a_{33} & a_{34} & 0 \\ a_{41} & a_{42} & a_{43} & a_{44} & 0 \\ a_{51} & a_{52} & a_{53} & a_{54} & 0 \end{vmatrix}. \quad (3.9)$$

Here we have $a_{1j} = \beta_j + (\tau/2) + \kappa_0$, $a_{2j} = \xi_j(\beta_j - (\tau/2) + \kappa_0)$, $a_{3j} = \xi_j^*(\beta_j + (\tau/2) - \kappa_0)$, $a_{4j} = \xi_j \xi_j^*(\beta_j - (\tau/2) - \kappa_0)$, and $a_{5j} = \xi_j(\beta_j - (\tau/2) - \kappa_0)$, where $\xi_j^* = \exp\{i[\beta_j - (\tau/2)]L\}$ is the thickness of the crystal, and the letter D denotes the determinant outlined by the dotted lines in (3.9). Expressions for the remaining amplitudes of the circular waves in (3.8) are also given by Eq. (3.9) by replacing in it the fifth row of the determinant. In order

to derive E_-^r , we must replace the components a_{5j} of the fifth row by $\beta_j + (\tau/2) - \kappa_0$; in order to get E_+^t , replace a_{5j} by $\xi_j^*(-\beta_j - (\tau/2) - \kappa_0)$; in order to get E_-^t , replace a_{5j} by $\xi_j \xi_j^*(-\beta_j + (\tau/2) - \kappa_0)$.

In addition to diffractive reflection, the expressions (3.8) and (3.9) take into account reflection by the boundaries of the CLC. A detailed analysis of the effect of reflection at the boundaries has been made in Refs. 28–30. At normal incidence, the reflection from the boundaries can prove substantial either in very thin specimens or far from the region of selective reflection. However, to take reflections into account quantitatively actually requires numerical calculations.

If we neglect the reflection from the boundaries, which is usually not very substantial, and allow for the fact that usually $\delta \lesssim 0.1$ for CLCs, then we get from (3.9) the simpler expressions that will be analyzed below.

c) The reflection coefficient

Upon solving the boundary problem in this manner (while treating the cholesteric helix as being right-handed for the sake of concreteness), we see that a levopolarized wave "excites" in the crystal only solution 1 or 4 (depending on the side of the crystal on which it is incident), and the latter penetrates the CLC without undergoing selective reflection. A dextropolarized wave near the frequency ω_B excites in the crystal the two eigenwaves 2 and 3. It undergoes reflection from the crystal, and the squared ratio of the amplitude of the reflected wave E_+^r to that of the incident wave E_+^i is given by the following expression:

$$R_+ = \left| \frac{E_+^r}{E_+^i} \right|^2 = \frac{\kappa^2 \delta^2 \sin^2 \beta_3 L}{\tau^2 \beta_3^2 + \kappa^2 \delta^2 \sin^2 \beta_3 L}. \quad (3.10)$$

Here we have $\beta_3^2 = \kappa^2 + (\tau^2/4) - \kappa \sqrt{\tau^2 + \kappa^2 \delta^2}$.

We note that the selective reflection by the CLC of dextropolarized light leads to right-circular polarization also in the reflected wave, whereas the circular polarization reverses sign upon reflection from ordinary materials (i.e., in nondiffractive reflection).

Let us examine Eq. (3.10) in greater detail. In the frequency range $\omega_B/\sqrt{1+\delta} < \omega < \omega_B/\sqrt{1-\delta}$ (i.e., when $|\kappa^2 - \tau^2/4| < \kappa^2 \delta$), the quantity β_3 becomes imaginary. Hence $\sin \beta_3 L$ in (3.10) transforms into $i \sinh |\beta_3| L$. At a great enough thickness of the crystal such that $L \gg p/\delta\pi$, we have $\sinh |\beta_3| L \gg 1$, and hence $R_+ \approx 1$. That is, total selective reflection of right-circularly polarized light occurs (Fig. 5). The frequency width of the reflection region is $\Delta\omega = \omega_B/\sqrt{1-\delta} - \omega_B/\sqrt{1+\delta} \approx \delta\omega_B$. That is, it is determined by the anisotropy of the dielectric permittivity δ . Outside the region of selective reflection, the intensity of the reflected light does not decline smoothly as the frequency leaves the region of strong reflection, but according to (3.10), it oscillates. These oscillations stem from the diffraction of light in the limited volume, and do not involve reflection from the boundaries. Usually these oscillations are not observed experimentally owing to imperfection of the specimens.

In the case of thin crystals ($L \ll p/\delta\pi$) we have

$$R_+ \approx \delta^2 \frac{\sin^2 [\kappa - (\tau/2)] L}{[(2\kappa - \tau)/\tau]^2} \quad (3.11)$$

That is, the intensity of the reflected light is proportional to δ^2 , while the reflection curve is greatly broadened (see Fig. 5). This result can also be derived from the formula (2.3) of the kinematical approximation. Figure 5 also shows the intermediate case $L \sim p/\pi\delta$. At the characteristic values of $p \sim 6000 \text{ \AA}$, $\delta \sim 0.02$, we have $p/\pi\delta \sim 10 \text{ \mu m}$. That is, both limiting cases of $L \gtrless p/\pi\delta$ can be realized experimentally. For the polarization n_+ , the intensity of the wave $|E_+^t|^2$ transmitted by the crystal and the transmission coefficient T_+ are determined by the expression

$$T_+ = \left| \frac{E_+^t}{E_+^i} \right|^2 = \frac{\tau^2 \beta_3^2}{\tau^2 \beta_3^2 + \kappa^4 \delta^2 \sin^2 \beta_3 L} \quad (3.12)$$

Experimentally it is often convenient to measure the difference between the transmission coefficients of the two circular polarizations, i.e., the circular dichroism $D = (T_- - T_+)/ (T_- + T_+)$, where T_- is the transmission coefficient for the polarization n_- . The expression for D for nonabsorbing crystals can be written in the form

$$D = \frac{1 - T_+}{1 + T_+} = \frac{\kappa^4 \delta^2 \sin^2 \beta_3 L}{2\tau^2 \beta_3^2 + \kappa^4 \delta^2 \sin^2 \beta_3 L} \quad (3.13)$$

As the presented formulas imply, in a nonabsorbing CLC the circular dichroism and its frequency-dependence stem from diffraction of light and are determined by the structural parameters of the CLC and do not involve directly the intrinsic dichroism of the molecules. However, for light frequencies in the region of an absorption band of the molecules, the effect of the intrinsic dichroism of the molecules can be quite substantial and is treated in the section on absorbing CLCs.

d) Rotation of the plane of polarization

The rotation of the plane of polarization in the medium arises from the difference of phase velocities of the waves having right- and left-hand circular polarization. Owing to its diffractive nature, the rotation of the plane of polarization in a CLC in the region of selective reflection proves incomparably larger than the "intrinsic rotation," which arises simply from the optical activity of the molecules of the CLC, and it has a number of qualitative peculiarities.

One of them involves the fact that the right circularly polarized wave is strongly attenuated in the CLC by diffractive reflection. Hence the light transmitted by the crystal varies in polarization, and has an elliptical rather than linear polarization that depends on the thickness of the specimen. In this connection we should speak of the rotation, say, of the major axis of the ellipse of polarization. The ratio of the axes of the ellipse of polarization in the wave transmitted through the specimen is fully determined by the following expression (under the condition that the light incident on the crystal is linearly polarized):

$$b = \frac{1 - \sqrt{T_+}}{1 + \sqrt{T_+}} \quad (3.14)$$

Here T_+ is given by (3.12).

Another peculiarity is manifested in the fact that the

rotation of the plane of polarization depends very strongly on the frequency, even far from absorption bands, and the direction of rotation proves to be different on different sides of the Bragg frequency ω_B . For the frequency ω_B itself the rotation of the plane of polarization vanishes. This frequency-dependence of the magnitude and sign of the rotation has a simple, natural explanation. Of the two eigenwaves that correspond to the circular polarization that is diffracted by the structure of the CLC and which are represented in the expansion (3.7), one has a smaller phase velocity than the wave not diffracted by the CLC, while the other has a greater velocity. Depending on the sign of the frequency detuning of the incident wave with respect to ω_B in the crystal, either the "slow" or the "fast" diffracted wave is mainly excited. This leads to the observed frequency-dependence of the magnitude and sign of the rotation. And finally, let us mention another unusual property that distinguishes a CLC from ordinary gyrotropic media: the angle of rotation of the plane of polarization (or more exactly, the axis of the ellipse of polarization) depends nonlinearly on the thickness of the specimen.

We find from the solution of the boundary problem that the amplitude of the wave emerging from the crystal is given by the following expression (the incident wave is linearly polarized):

$$\vec{E}^t = \frac{E^i}{\sqrt{2}} \left\{ n_- e^{i(\beta_1 - \tau/2)L} + n_+ \frac{\beta_3 \tau e^{i\tau L/2}}{\beta_3 \tau \cos \beta_3 L + i \{ \beta_3^2 + (\tau/2)^2 - \kappa^2 \} \sin \beta_3 L} \right\} \quad (3.15)$$

Here β_1 and β_3 are defined in (3.5).

The phase difference of the two terms in (3.15) determines the angle of rotation φ of the plane of polarization:

$$\varphi = \frac{1}{2} \left\{ (\beta_1 - \tau) L + \arctg \left[\frac{\beta_3^2 + (\tau/2)^2 - \kappa^2}{\beta_3 \tau} \operatorname{tg} \beta_3 L \right] \right\} \quad (3.16)$$

The expression (3.16) is simplified for crystals of great thickness:

$$\frac{\varphi}{L} \approx \operatorname{Re} \frac{\beta_1 - \beta_3 - \tau}{2} \quad (3.17)$$

Figure 6 shows the frequency dependences of the rotation of the plane of polarization as described by Eqs. (3.16) and (3.17). Far from the region of selective reflection, Eqs. (3.16) and (3.17) go over into the well-known formula of De Vries²⁶:

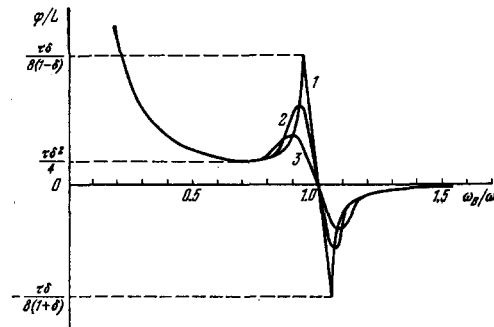


FIG. 6. Frequency dependence of the rotation angle of the plane of polarization for specimens of varying thicknesses. Curve 1—for $L\delta/p \gg 1$, 2—for $L\delta/p = 1$, 3—for $L\delta/p = 0.5$; the curves are drawn for $\delta \approx 0.1$.

$$\frac{\varphi}{L} \approx \frac{\kappa^2 \delta^2}{\tau(4\kappa^2 - \tau^2)} \quad (3.18)$$

The formula of De Vries has the defect that φ/L approaches $\pm\infty$ in the region of selective reflection, which does not happen in the exact expression (3.16).

e) Rotation of the plane of polarization in thin layers

Whenever the reflection from the crystal is not too great (and it is precisely these crystals that are usually employed for observing rotation), an approximate expression is obtained from (3.16) that functions well both in the region of selective reflection and far from it:

$$\frac{\varphi}{L} \approx \frac{\kappa^2 \delta^2}{4\tau^2} \left[-\frac{1}{\kappa + (\tau/2)} + \frac{\kappa - (\tau/2)}{\beta_3^2} \left(1 - \frac{\sin 2\beta_3 L}{2\beta_3 L} \right) \right] \quad (3.19)$$

For cruder calculations we can assume that $\beta_3 \approx \kappa - (\tau/2)$ and derive from (3.19) a quite simple expression:

$$\frac{\varphi}{L} \approx \frac{\kappa^2 \delta^2}{\tau(4\kappa^2 - \tau^2)} - \frac{\kappa^2 \delta^2 \sin(2\kappa - \tau)L}{2\tau^2(2\kappa - \tau)^2 L} \quad (3.20)$$

The first term in (3.20) coincides with the expression of De Vries (3.18); the second term in (3.20) is substantial only in the region of selective reflection, as $\kappa \rightarrow \tau/2$. On the whole they give the correct variation of the rotation angle for all frequencies. Quantitatively the expressions (3.19) and (3.20) differ little from the exact expression (3.16) when the thickness of the specimen is small such that $L < p/\pi\delta$. Actually one can apply Eqs. (3.19) and (3.20) when the rotation angle is restricted by $\varphi < 1$. We note again that Eqs. (3.16), (3.19) and (3.20) yield a nonlinear relationship between the rotation angle φ and the thickness L in the region of selective reflection. Thus, at small L near the maxima of rotation (see Fig. 6), we have $\varphi \sim L^2$, while near the frequency that corresponds to change of sign of the rotation, we have $\varphi \sim L^3$.

f) The limit $\lambda \ll p$

In this case only the waves 1 and 3 (or 2 and 4) are excited in the crystal, depending on the direction of propagation of the incident wave. We obtain from the formulas (3.9) the following expression for the amplitude of the transmitted wave (3.8):

$$\begin{aligned} E_+^t &= \left[E_+^i \left(\cos \frac{\beta_1 - \beta_3}{2} L - it \sin \frac{\beta_1 - \beta_3}{2} L \right) \right. \\ &\quad \left. + E_{-i}^i \frac{\kappa \delta}{\tau} \sin \frac{\beta_1 - \beta_3}{2} L \right] \exp \left[i(\beta_1 + \beta_3 + \tau) \frac{L}{2} \right], \\ E_-^t &= \left[E_-^i \left(\cos \frac{\beta_1 - \beta_3}{2} L + it \sin \frac{\beta_1 - \beta_3}{2} L \right) \right. \\ &\quad \left. + E_{+i}^i \frac{\kappa \delta}{\tau} \sin \frac{\beta_1 - \beta_3}{2} L \right] \exp \left[i(\beta_1 + \beta_3 - \tau) \frac{L}{2} \right]. \end{aligned} \quad (3.21)$$

Here we have $t = 1/\sqrt{1 + (\kappa^2 \delta^2 / \tau^2)}$. The rotation angle φ varies nonlinearly with the thickness of the crystal and it generally depends on the direction of the initial polarization.

Two cases can occur under the adopted restrictions on κ . If $\kappa \ll \tau/\delta$, then Eq. (3.21) yields (3.18), and then the angle of rotation does not depend on the initial polarization.

In the limiting case $\kappa \gg \tau/\delta$, i.e., at very short wavelengths, the results depend on the orientation of the molecules at the surface of the CLC. In the general case Eq. (3.21) implies that linearly polarized incident light yields elliptically polarized light at the exit. If

the plane of polarization of the incident light is parallel (or perpendicular) to the orientation of the CLC molecules at the entrance surface, then the emerging light is linearly polarized and its plane of polarization is also parallel (or perpendicular) to the orientation of the molecules at the exit surface. Thus the plane of polarization seems to "track" the orientation of the molecules of the CLC and the angle φ is determined by the number of turns of the cholesteric helix, i.e., $\varphi = 2\pi L/p$. For optical wavelengths, the relationship $\kappa \gg \tau/\delta$ can be realized in mixtures of dextro- and levorotatory CLCs,³¹ and also in twisted nematic twist-cells;^{5,13} this case was first studied by Mauguin.³²

We have studied above the optical properties of CLCs for the propagation of light along the axis of the helix^{21,22,26,27} (see also Refs. 33–38 and 146, in which an exact solution in a magnetic field was found). In a number of studies by Chandrasekhar and other authors,^{39–41} this same problem has been solved by approximate methods. The results obtained by the approximate methods were close to the exact results for small δ .^{8,42}

The case of normal incidence of light on a CLC has been studied in detail experimentally. The results of the experimental studies agree well with the theoretical conclusions.

In Refs. 40 and 43–51 reflection of light from a CLC and circular dichroism were studied. The results of these studies confirm the diffractive nature of the reflection and the circular dichroism in a CLC. In particular, this is implied by the observed dependence of the reflection and the dichroism on the frequency of the light and the thickness of the specimen^{46,47,50} (Fig. 5). In Fig. 7a we see also the theoretically implied diffractive beating of the reflection coefficient with the frequency at the edges of the reflection curve.

The rotation of the plane of polarization in a CLC has been studied in a large number of investigations, both in the region of selective reflection^{43,44,46,49,53–55} and

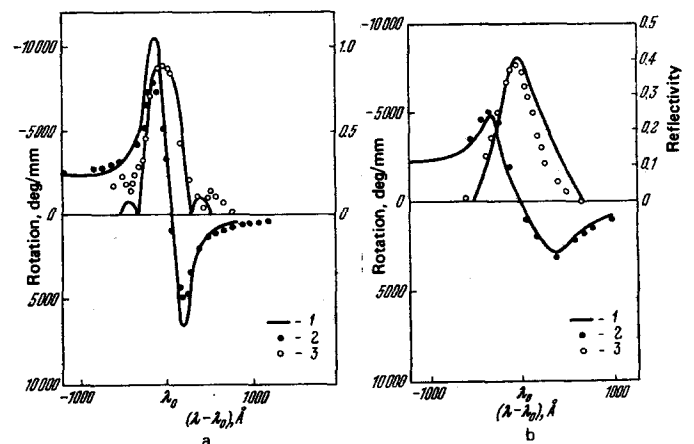


FIG. 7. Calculated and measured values of the optical rotatory dispersion and reflectivity for a specimen of cholesteryl cinnamate 3- μm (a) and 1- μm (b) thick.⁴⁶ 1—theory; 2—experiment for optical rotatory dispersion; 3—experiment for reflectivity.

outside this region⁵⁶⁻⁶² (see Fig. 7). The results of the measurements agree well with the theory, and as Sonin and his coauthors have shown,⁵⁴ they can be used to determine the parameters of a CLC and their temperature dependences.

The above-noted nonlinear dependence of the rotation of the plane of polarization on the thickness of the specimen is distinctly manifested experimentally (cf. Figs. 7a, b).

We note also that the circular dichroism and the rotation of the plane of polarization, which are measured experimentally as independent quantities, are linked by dispersion relations.^{63,64} This means that a knowledge throughout the entire frequency interval of one of the quantities allows us in principle to determine the other one. Therefore it seems useful to employ these relations for testing the consistency of measurements of the rotation and the circular dichroism. In this connection the as yet unstudied problem of the dispersion relations under conditions of optical activity of a diffractive nature would be interesting to study theoretically and experimentally.

4. OBLIQUE INCIDENCE OF LIGHT

This case is more complicated to analyze since an exact solution of Maxwell's equations is not known here. Hence a number of studies have performed a general analysis of the structure and properties of the solutions.^{21,22,65-67} However, numerical methods⁶⁸⁻⁷¹ have been employed to get solutions for concrete values of the parameters of the problem. Yet the existence of a small parameter (the anisotropy δ of the dielectric properties) allows one to develop a rather accurate approximate theory.^{27,72-74}

It has been shown that the case of oblique incidence of light manifests qualitative differences in the optics of CLCs. Higher orders of reflection exist at frequencies that are multiples of the Bragg frequency. The first-order reflection is also substantially changed: a region of reflection of light of any polarization appears, and the polarization properties of the solutions become complicated. The physical source of these peculiarities is that one must take into account the double refraction of light in describing the propagation of light at an angle to the axis of the helix, in addition to the diffractive scattering of light with the polarization defined by (2.8). In particular, the birefringence exerts an effect on the polarization characteristics of the solutions. The existence of diffractive scattering of any polarization leads to a more complicated pattern of the diffraction of light by a CLC in the case of oblique incidence than at normal incidence.

The results obtained by the approximate methods^{27,72-74} are essentially analogous and they agree with the results of the general analysis. Yet they differ from them in greater simplicity, and they yield an analytic description of the problem. Hence we shall present below the case of oblique incidence, mainly following Refs. 72-74, which have applied the methods of the dynamical theory of diffraction⁷⁵⁻⁷⁷ to describe

the optical properties of CLCs.

a) System of dynamical equations

The equation for the electric field vector in the case of arbitrary direction of propagation of light in a CLC has the form

$$\hat{\epsilon} \frac{\partial^2 \vec{E}}{\partial t^2} = -c^2 \text{rot rot } \vec{E}. \quad (4.1)$$

Here $\hat{\epsilon}$ is given by Eq. (1.3).

Owing to the periodicity of the CLC, the solution of Eq. (4.1) has the form of Bloch waves:

$$\vec{E}(\mathbf{r}, t) = \exp(i\mathbf{k}_0 \mathbf{r} - \omega t) \sum_{s=-\infty}^{\infty} \vec{E}_s \exp(is\tau \mathbf{r}). \quad (4.2)$$

Upon substituting (4.2) into (4.1), we get a homogeneous system of equations for determining \vec{E}_s :

$$-\mathbf{k}_s^2 \vec{E}_s + \frac{\omega^2}{c^2} \sum_{s'=-\infty}^{\infty} \hat{\epsilon}_{s-s'} \vec{E}_{s'} + (\kappa_s \vec{E}_s) \mathbf{k}_s = 0. \quad (4.3)$$

Here we have $\mathbf{k}_s = \mathbf{k}_0 + s\tau$, while the $\hat{\epsilon}_s$ have been defined in (2.2). The general properties of Eq. (4.1) have been analyzed by Dreher and Meier,⁶⁷ who showed by using the theorem of Floquet (see Ref. 78) that four values of \mathbf{k}_0 exist for a fixed frequency and angle of incidence in the crystal at which Eq. (4.1) has a non-trivial solution of the form (4.2).

Application of numerical methods for solving the system (4.3)⁶⁸ allows one to get its solution to any degree of accuracy. However, it is more convenient to have an analytical expression for analyzing the properties of the solutions. Hence we shall use below in solving (4.3) the two-wave approximation of the dynamical theory of diffraction that is well known from the electrodynamics of periodic media and x-ray diffraction.^{75-77,79} This approximation employs the fact that only two amplitudes \vec{E}_0 and \vec{E}_s have a large value in the system (4.3) when the Bragg condition (2.4) is satisfied. They are of the order of the amplitude of the wave incident on the crystal, while the others are smaller by at least a factor of δ .

b) First-order diffractive reflection

Let us examine the first-order reflection, $s=1$. Let us isolate from the system (4.3) the two vector equations for the waves \vec{E}_0 and \vec{E}_1 having the wave vectors \mathbf{k}_0 and $\mathbf{k}_1 = \mathbf{k}_0 + \tau$:

$$\begin{aligned} \left(\hat{\epsilon}_0 - \frac{\mathbf{k}_0^2 c^2}{\omega^2} \right) \vec{E}_0 + \hat{\epsilon}_{-1} \vec{E}_1 &= 0, \\ \hat{\epsilon}_1 \vec{E}_0 + \left(\hat{\epsilon}_0 - \frac{\mathbf{k}_1^2 c^2}{\omega^2} \right) \vec{E}_1 &= 0. \end{aligned} \quad (4.4)$$

The accuracy of the approximation employed has allowed us to neglect in (4.4) the small departure from transverseness of the waves \vec{E}_0 and \vec{E}_1 , which is of the order of δ , and to consider \vec{E}_0 and \vec{E}_1 to be respectively orthogonal to \mathbf{k}_0 and \mathbf{k}_1 .

In order to solve the system (4.4), it is convenient to reduce it to the form that is usually employed in the dynamical theory of diffraction⁷⁷:

$$\left. \begin{aligned} \left(1 - \frac{k_0^2}{\kappa^2} + \frac{\delta}{2} \cos^2 \theta\right) E_0^\sigma - \frac{\delta}{2} E_1^\sigma + \frac{i\delta}{2} \sin \theta \cdot E_1^\pi &= 0, \\ \left(1 - \frac{k_1^2}{\kappa^2} - \frac{\delta}{2} \cos^2 \theta\right) E_0^\pi - \frac{i\delta}{2} \sin \theta E_1^\sigma - \frac{\delta}{2} \sin^2 \theta \cdot E_1^\pi &= 0, \\ -\frac{\delta}{2} E_0^\sigma + \frac{i\delta}{2} \sin \theta \cdot E_0^\pi + \left(1 - \frac{k_1^2}{\kappa^2} + \frac{\delta}{2} \cos^2 \theta\right) E_1^\sigma &= 0, \\ -\frac{i\delta}{2} \sin \theta E_0^\sigma - \frac{\delta}{2} \sin^2 \theta \cdot E_0^\pi + \left(1 - \frac{k_1^2}{\kappa^2} - \frac{\delta}{2} \cos^2 \theta\right) E_1^\pi &= 0. \end{aligned} \right\} \quad (4.5)$$

Here E^σ and E^π are the σ - and π -components of the amplitudes of E_0 and E_1 , and

$$\kappa = \frac{\omega}{c} \sqrt{\varepsilon \left(1 - \frac{\delta}{2} \cos^2 \theta\right)}$$

is the mean value of the wave vector of the wave propagating in the CLC at an angle of $\pi/2 - \theta$ to the optic axis.

Upon equating the determinant of the system (4.5) to zero, we obtain the condition for its solvability in a form analogous to (3.4):

$$q^4 - (2\Delta^2 + 2m^2 - 1)q^2 + \Delta^4 - (2m^2 + 1)\Delta^2 + 2m^2\Delta = 0. \quad (4.6)$$

Here we have introduced the following notation:

$$\begin{aligned} q &= \frac{k_1^2 - k_0^2}{\kappa^2 \delta (1 + \sin^2 \theta)}, \quad m = \frac{\cos^2 \theta}{1 + \sin^2 \theta}, \\ \Delta &= \frac{2\kappa^2 - k_0^2 - k_1^2}{\kappa^2 \delta (1 + \sin^2 \theta)}. \end{aligned} \quad (4.7)$$

c) Eigensolutions

The relationships (4.6) and (4.7) define the region of values of the wave vectors k_0 and k_1 compatible with the Bragg condition (2.4) that satisfy the Maxwell equations. These regions form in k -space the so-called dispersion surfaces.^{75,76,79} Without stopping to analyze the dispersion surfaces, below we shall give directly the solutions that satisfy the boundary conditions.

In the case in which the optic axis is perpendicular to the surface of the CLC, the parameter Δ is associated with the deviation of the angle of incidence or the wavelength of the light from the values defined by the Bragg condition (2.4). It can be represented in the form

$$\Delta = \frac{2(2\kappa \sin \theta - \tau) \sin \theta}{\kappa \delta (1 + \sin^2 \theta)}. \quad (4.8)$$

In particular, for a fixed angle θ , we have $\Delta = 4(\omega - \omega_B) \sin^2 \theta / \omega \delta (1 + \sin^2 \theta)$, where $\omega_B = \tau c / 2 \sin \theta \sqrt{\varepsilon (1 - (\delta/2) \cos^2 \theta)}$. For a fixed frequency ω , we have $\Delta \approx 2(\theta - \theta_B) \sin 2\theta_B / \delta (1 + \sin^2 \theta_B)$, where $\sin \theta_B = \tau / 2\kappa$.

The quantity q , which is determined from Eq. (4.6), describes the change in the wave vector caused by diffraction and is analogous to β in (3.4).

Since Eq. (4.6) is biquadratic, we can easily find its roots q_j ($j=1, 2, 3, 4$) that correspond to the eigensolutions E_{0j} and E_{1j} of the systems (4.4) and (4.5), and hence find the solutions of Eq. (4.1):

$$\vec{E}_j(r, t) = (E_{0j} e^{ik_0 r} + E_{1j} e^{ik_1 r}) e^{-i\omega t}. \quad (4.9)$$

Here we have

$$\left. \begin{aligned} E_{0j} &= E_{0j}^\sigma \sigma + E_{0j}^\pi \pi, \quad E_{1j} = E_{1j}^\sigma \sigma + E_{1j}^\pi \pi, \\ E_{0j}^\sigma &= (\Delta - m + q_j) [(\Delta - q_j)^2 - m^2], \\ E_{0j}^\pi &= -i \sin \theta_B (\Delta + m + q_j) [(\Delta - q_j)^2 - m^2], \\ E_{1j}^\sigma &= (\Delta - m - q_j) (\Delta + q_j - m^2), \\ E_{1j}^\pi &= -i \sin \theta_B (\Delta + m - q_j) (\Delta + q_j - m^2), \end{aligned} \right\} \quad (4.10)$$

$$k_{0j} = \kappa_0 + n \frac{\kappa \delta (\Delta + m + q_j)}{2 \sin \theta (1 + m)}, \quad k_{1j} = k_{0j} + \tau. \quad (4.11)$$

Eq. (4.11) has been derived by employing the boundary conditions on the wave vectors (continuity of their tangential components) for the case where the surface of the specimen is perpendicular to the optic axis of the CLC. In (4.11) κ_0 is the wave vector of the wave incident on the CLC while outside the specimen and n is the normal to the surface of the crystal.

The eigensolutions (4.9) amount to superposition of two waves having the wave vectors k_{0j} and k_{1j} , which are generally elliptically polarized. The general solution of Eq. (4.1) is a superposition of the eigensolutions of (4.9):

$$\vec{E}(r, t) = \sum_{j=1}^4 C_j \vec{E}_j(r, t). \quad (4.12)$$

Here the coefficients C_j are determined from boundary conditions analogous to the case of normal incidence. Having found $\vec{E}(r, t)$ in the crystal, we can easily also find the waves emerging from the crystal, reflected and transmitted. Under the assumption that the reflection at the boundaries arising from the refractive-index difference is small, a solution of this problem has been given in Ref. 73 (see also Ref. 77). Here we shall merely discuss the obtained results.

If the crystal is thin enough ($L \ll p/\delta$), then naturally we get results that coincide with the kinematical approximation. Namely, only the wave having the polarization defined by (2.8) is reflected, and the reflected wave has the same polarization.

d) Thick crystals

For thick crystals ($L \gg p/\delta$), the solution of the boundary problem is simplified in comparison with the case $L \sim p/\delta$. This is because in this case only the eigensolutions that correspond to attenuation of the wave as it propagates into the interior of the crystal have nonzero coefficients in the superposition (4.12). The attenuation of the eigensolutions (for real ε) arises from diffractive scattering, and it corresponds to the existence of nonzero imaginary components in the expressions (4.11) for k_0 and k_1 in certain ranges of values of the parameter Δ . The latter implies the existence in k -space of bands forbidden for propagating waves. In turn this leads to the appearance of bands of diffractive reflection.^{67,68,72,73} The boundaries of these bands are most simply determined from Eq. (4.6).

As Eq. (4.6) implies, we can distinguish three different cases in the reflection of light from a CLC, depending on the deviation (in angle of incidence or frequency) from the Bragg condition (2.4), i.e., depending on the parameter Δ of (4.8):

1) If Δ has a value such that all solutions q_j of Eq. (4.6) are real, then the wave vectors (4.11) of all the solutions of (4.9) are also real. These waves pass through the crystal without attenuation, and the reflection coefficient in this region is small.

2) Another characteristic case involves values of Δ for which two solutions $q_{1,2}$ are imaginary (differing only in sign), while the other two $q_{3,4}$ are real. We see

from (4.9) that in this case one of the eigenwaves (say, \vec{E}_1) decays exponentially into the interior of the crystal, while the other (\vec{E}_2) increases. Here a wave incident on the crystal with an elliptical polarization corresponding to E_{01} is totally reflected, while the wave with orthogonal polarization excites in the crystal the nondecaying waves \vec{E}_3 and \vec{E}_4 and contributes very weakly to the reflection. For this region of Δ values the reflection coefficient for unpolarized light is practically 1/2.

3) There is another region of values of the parameter Δ in which all four roots q_i are complex (complex conjugates in pairs). In this region light of any polarization is practically completely reflected, and the reflection coefficient for unpolarized light is unity.

The polarization properties of reflection in CLCs are described in greater detail in Ref. 73. As has been shown,^{67,68,72,73} the angular (frequency) regions of selective reflection of a particular polarization and of reflection of light of any polarization can either adjoin one another or be separated by regions of weak reflection. Figure 8 illustrates qualitatively the nature of the reflection of unpolarized light. The reflection coefficient 1/2 corresponds to the region of reflection of one polarization state, which varies along the reflection curve; the value unity corresponds to the region of reflection of any polarization. At small angles of reflection θ , the regions of total and selective reflection may be not adjoining one another, and be separated by intervals of very weak reflection. In particular, this is implied by Fig. 9, which shows the structure of the reflection bands as derived from (4.6) as a function of the wavelength and the angle of incidence on the crystal.^{72,73} We see that at normal incidence ($\theta = 90^\circ$) the width of the region of reflection of light of any polarization vanishes, in agreement with the results of the exact solution.

The above-presented analytical description agrees with the results of numerical calculations and experimental studies of the optical characteristics of CLCs with oblique incidence. Berreman and Scheffer⁶⁹⁻⁷¹ (Fig. 10) have demonstrated the agreement of numerical calculations with the results of optical measurements on cholesteric-nematic mixtures. In particular, attention is called to the appearance of the higher orders of reflection with oblique incidence of light.

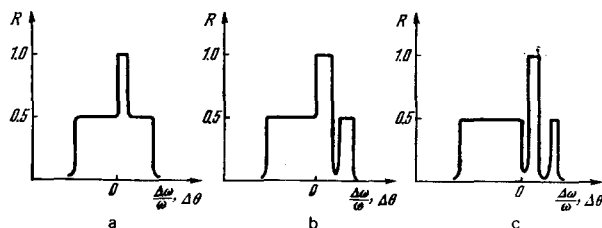


FIG. 8. Qualitative form of the reflection coefficient R for unpolarized light as a function of the deviation in frequency (or angle) from the Bragg condition. Depending on the angle of incidence θ , there are: (a) one ($90^\circ > \theta \geq 32^\circ$), (b) two ($32^\circ \geq \theta \geq 25^\circ$), or (c) three ($25^\circ \geq \theta > 0$) regions of selective reflection.

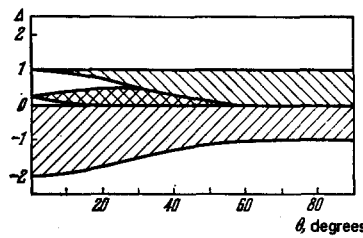


FIG. 9. Regions of diffractive reflection as a function of the angle of incidence of light on a CLC. The parameter Δ (see (4.8)) characterizes the deviation from the Bragg condition. The single hatching shows the regions of selective reflection of one polarization, and the double hatching the region of reflection of any polarization.

e) Higher orders of reflection

Let us examine another feature of the case of oblique incidence—the existence of higher orders of reflection, i.e., the existence of diffractive reflection in CLCs not only for $s = 1$ in Eq. (2.4), but also for $s > 1$. At a fixed angle of incidence, the higher orders are manifested in the existence of diffractive reflection of light, not only in the frequency region of ω_B , but also in the regions $s\omega_B$, where s is the order of reflection.

The higher orders of reflection were first obtained theoretically, by a numerical solution of Maxwell's equations,⁶⁸⁻⁷¹ while they were also observed experimentally in Refs. 69-71.

As we have noted, reflection at frequencies that are multiples of ω_B is absent at normal incidence. At oblique incidence, as the results of the kinematical approximation show, direct scattering of light with change of the wave vector by $s\tau$ also cannot occur if $s > 1$, since the corresponding harmonics are absent in the Fourier expansion (2.2) of the tensor $\hat{\epsilon}$. However, if we use the dynamical theory of diffraction, in particular Eq. (4.3), we can not only show that the higher orders of reflection exist at oblique incidence, but can analyze their dependence on s . Here it turns out that the "strength" of diffractive reflection rapidly declines with increasing order s of reflection.

In spite of the fact that there is no direct scattering

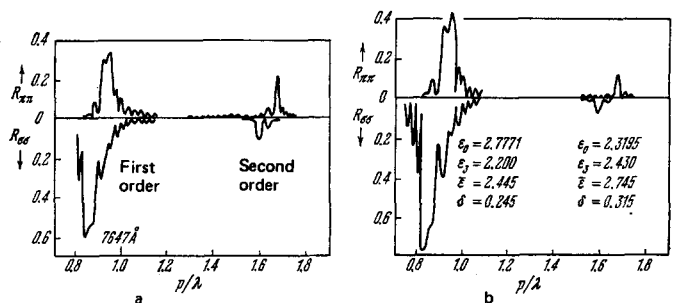


FIG. 10. Measured (a) and calculated (b) frequency- and polarization-dependences in the first and second orders of reflection for a two-component cholesteric specimen.⁷⁰ $R_{\pi\pi}$ ($R_{\sigma\sigma}$) is the reflection coefficient for (π) (σ) polarization into (π) (σ) polarization; the angle of incidence is 45° ; the specimen thickness is $L = 11.47 \mu\text{m}$; we have $L/p = 15$; and ϵ_0 is the dielectric permittivity of the medium outside the specimen.

of the waves E_0 and E_s , nevertheless such scattering proves possible via the intermediate waves E_1, \dots, E_{s-1} .⁷⁴ In order to describe these processes, we must keep in the system (4.3) at least $s+1$ equations containing the amplitudes of the intermediate waves. These amplitudes E_1, \dots, E_{s-1} are smaller by at least a factor of δ than E_0 and E_s . We can eliminate them from (4.3) by expressing them in terms of E_0 and E_s . Consequently we get a system of equations for E_0 and E_s that is analogous to (4.4), but with $\hat{F}_s \epsilon_{s+1}$ substituted for $\hat{\epsilon}_{s+1}$, where we have

$$F_s = \left(\frac{\delta}{8}\right)^{s-1} \frac{s^{2s}}{(s!)^2} (\cot \theta)^{2s-2}. \quad (4.13)$$

Just as the quantity δ does in the first order of reflection, now it is $F_s \delta$ that determines the characteristic intervals of angles (or frequencies) of diffractive reflection, which prove to be of the order of δ^s . That is, they decline rapidly with increasing s . We should also note that one needs a rather thick crystal ($L \sim p/\delta F_s$) to observe reflections of the s th order, and actually one can speak of observing them only at not too large s .

Figure 11 shows the frequency-dependence of the reflection coefficient for unpolarized light for the second order of reflection.⁷⁴ The two side maxima are separated from the central one by distances of $\pm(\omega\delta/4)\cot^2\theta$; θ is the angle of incidence on the crystal. The width of the σ -maximum is $\Delta\omega_\sigma = \omega\delta^2 \cos^2\theta/4 \sin^4\theta$. In it, light with linear σ -polarization is also reflected as σ -polarized light. In the π -maximum, π -polarized light is reflected as π -polarized; its width is $\Delta\omega_\pi = (\omega\delta^2/4)\cot^2\theta$. In the central maximum, light of any polarization is reflected. Here σ -polarization gives π -polarization on reflection, and vice versa. The width of this maximum is $\Delta\omega_T = (\omega\delta^2/4)\cos^2\theta/\sin^2\theta$. If the thickness L of the crystal is small, the maxima in Fig. 11 are broadened (as shown by the dotted line) and they become poorly marked (as we can also see in Fig. 10).

One can also observe the same pattern of three maxima upon varying the angle of incidence at constant frequency. The second order of reflection occurs near the Bragg angle $\theta_B = \arcsin(\tau/\kappa)$, while the angular widths of the σ -, π -, and T -maxima are determined as

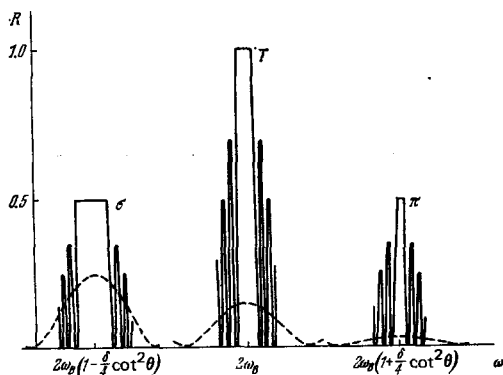


FIG. 11. Calculated frequency dependence of the reflection coefficient R for unpolarized light in the second order.⁷⁴ σ — σ -polarization is reflected; T —total reflection of any polarization, π — π -polarization is reflected (solid curve—for a thick specimen; dotted—for a thin one).

follows:

$$\frac{\Delta\theta_\sigma}{\Delta\omega_\sigma} = \frac{\Delta\theta_\pi}{\Delta\omega_\pi} = \frac{\Delta\theta_T}{\Delta\omega_T} = \frac{1}{\omega} \operatorname{tg} \theta. \quad (4.14)$$

No detailed experimental study of reflection of the second and higher orders has yet been made (apart from Refs. 69–71).

Studies of higher-order reflections (in particular, second-order) might be useful for studying cholesteric crystals having large helix periods for which the first order lies in the infrared frequency range. We note that the Bragg angles are small in these crystals. This simplifies observation of the higher-order reflections. Actually, as we see from the above-presented formulas, in this case the frequency widths, and hence also the angular widths, of the reflection regions are increased (and also the thickness of crystal needed for effective observation is diminished).

5. OPTICS OF CHOLESTERIC LIQUID CRYSTALS IN AN EXTERNAL FIELD

In connection with the widespread application of liquid crystals in electro(magneto)optic devices, it is of substantial interest to study the effect of external fields on the optical properties of CLCs. This effect is based on the anisotropy of the local dielectric (magnetic) susceptibility of CLCs. This gives rise to forces that distort the structure of the CLC upon application of the external field. And in turn the change in structural properties is reflected in the optical characteristics of the CLCs.

The manifestations of the action of external fields on the structure (and hence also on the optical properties) are highly varied. They depend on the relationship between the susceptibilities χ_{\parallel} along the long axis of the molecules and χ_{\perp} in the transverse direction, the orientation of the applied field, and the geometry of the experiment (the dimensions of the optical cell, the original configuration of the CLC, etc.^{5,13,80}). For example, when $\chi_{\perp} > \chi_{\parallel}$, a field parallel to the optic axis exerts a stabilizing influence on the structure of the CLC: an imperfect or even a polycrystalline specimen of a CLC can be converted by an external field into a plane configuration. If $\chi_{\perp} < \chi_{\parallel}$, then, as de Gennes⁸¹ and Meyer⁸² have shown, the cholesteric helix becomes distorted in a field perpendicular to the optic axis, and its pitch p increases with increasing field, while complete untwisting of the helix occurs when a certain critical field is reached.

The examples given above correspond to situations in which surface effects are inessential. However, in thin cholesteric layers (optical cells), one cannot neglect surface effects, whereby the behavior of the CLC in the field becomes complicated and is determined by the competition of the surface and the "field" forces. Thus, for example, when $\chi_{\perp} < \chi_{\parallel}$, application to a plane configuration of a field parallel to the optic axis of the CLC gives rise to two-dimensional deformations of the structure of the CLC that depend on the thickness of the specimen.⁸³ In speaking of the effect of a field on the optical properties, we have not specified whether this field is electric or magnetic,

since every magneto-optic phenomenon has its electro-optic analog. Yet we should bear in mind that a fully analogy between electro- and magneto-optic phenomena exists only when the electric fields do not give rise to currents in the CLC (see the recent reviews of Blinov and Belyaev^{13,80}). Electro-optic phenomena that involve flow of currents, e.g., dynamic light scattering, lack magnetic analogs.

a) Structure and dielectric properties of cholesteric liquid crystals in a field

We shall treat below the optical properties of a CLC having a positive magnetic or electric anisotropy ($\chi_{\parallel} > \chi_{\perp}$) is a field perpendicular to its optic axis. As has been shown in Refs. 81 and 82, for this direction of the external field, the rotation angle of the molecules in (1.1) ceases to be a linear function of the coordinate z . The function $\varphi(z)$ is transformed into the elliptic amplitude of Jacobi, and correspondingly the $\sin\varphi$ and $\cos\varphi$ in the formulas (1.3) for the tensor $\hat{\epsilon}(z)$ are replaced by the elliptic sine and cosine.⁸⁴ Consequently all the harmonics are present in the Fourier expansion of the tensor $\hat{\epsilon}(z)$:

$$\hat{\epsilon}(z) = \sum_{s=-\infty}^{\infty} \hat{\epsilon}_s \exp\left(-\frac{4\pi i s z}{p_F}\right). \quad (5.1)$$

Here $p_F = (2/\pi)^2 K(k)E(k)$ is the pitch of the helix in the external field F ; $K(k)$ and $E(k)$ are the complete elliptic integrals⁸⁴; and their modulus k is found from the equation $k/E(k) = F/F_c$, where F is the external field, and $F_c = (\pi^2/p)\sqrt{k_{22}/(\chi_{\parallel} - \chi_{\perp})}$ is the value of the critical field (magnetic or electric) that completely untwists the cholesteric helix; k_{22} is the torsional elastic constant. The Fourier components of $\hat{\epsilon}_s$ prove to be:

$$\hat{\epsilon}_0 = \begin{pmatrix} \bar{\epsilon} + \frac{\bar{\epsilon}\delta\rho_0}{2} & 0 & 0 \\ 0 & \bar{\epsilon} - \frac{\bar{\epsilon}\delta\rho_0}{2} & 0 \\ 0 & 0 & \epsilon_s \end{pmatrix}, \quad (5.2)$$

$$\hat{\epsilon}_{s>1} = \frac{\bar{\epsilon}\delta}{2} \begin{pmatrix} \rho_s & \mp i\rho'_s & 0 \\ \mp i\rho'_s & 1-\rho_s & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \hat{\epsilon}_{-s} = \hat{\epsilon}_s^*$$

where we have

$$\rho_0 = 4 \frac{K(k) - E(k)}{k^2 K(k)} - 2, \quad \rho_s = \frac{4\pi^2 \eta^s}{k^2 (1 - \eta^{2s}) K^2(k)}, \quad (5.3)$$

$$\rho'_s = \rho_s \frac{1 - \eta^{2s}}{1 + \eta^{2s}}, \quad \eta = \exp\left[-\frac{\pi K(\sqrt{1-k^2})}{K(k)}\right].$$

b) System of dynamical equations

One cannot solve exactly the wave equation (3.1) with the dielectric permittivity (5.1), even in the case of normal incidence. Some special solutions have been obtained⁸⁵⁻⁸⁷ by numerical methods. As above, we shall treat here the optics of the CLC in the external field on the basis of the two-wave approximation of dynamical diffraction theory,^{87,88} which allows us to obtain analytic expressions. Although this approach also permits us to treat the case of arbitrary angle of incidence of the light on the crystal,⁸⁸ we shall restrict the treatment here to presenting only the case of normal incidence, since here the external field alters the optical properties of the CLC most radically. In particular, the existence of all the Fourier components in the expansion of $\hat{\epsilon}$ gives rise to the higher orders of reflection even

with normal incidence.

The equations of the dynamical theory of diffraction in the presence of an external field have a form analogous to (4.4):

$$\left(\hat{\epsilon}_0 - \frac{k_0^2 c^2}{\omega^2}\right) E_0 + \hat{\epsilon}_{-1} E_s = 0, \quad (5.4)$$

$$\hat{\epsilon}_s E_0 + \left(\hat{\epsilon}_0 - \frac{k_s^2 c^2}{\omega^2}\right) E_s = 0.$$

Here $\hat{\epsilon}_0$ and $\hat{\epsilon}_{\pm s}$ are defined in (5.2). We obtain from the condition for solvability of the system (5.4) for the case of normal incidence a secular equation analogous to (4.6):

$$q^4 - 2(\rho_0^2 - \rho_s^2 - \rho'_s{}^2 + \Delta^2) q^2 + \Delta^4 - 2(\rho_0^2 + \rho_s^2 + \rho'_s{}^2) \Delta^2 + (\rho_0^2 - \rho_s^2 + \rho'_s{}^2)^2 = 0. \quad (5.5)$$

Here we have

$$q = \frac{k_0^2 - k_s^2}{k^2 \delta}, \quad \Delta = 4 \frac{\omega - s\omega_B}{\omega \delta}, \quad \omega_B = \frac{2\pi c}{p_F \sqrt{\epsilon}}$$

Thus the case of normal incidence in the presence of a field proves to resemble that of oblique incidence of light on a CLC in the absence of a field. Hence the analysis of the properties of selective reflection in the field is analogous to that conducted in Chap. 4.

In contrast to the case of normal incidence of light on a CLC undistorted by a field, the polarization characteristics of the scattering in the field prove to depend on the thickness of the specimen.

c) Reflection from thick crystals

Let us first treat the reflection of different orders from thick crystals. Upon application of the field, the region of selective first-order reflection of a circularly polarization state is shifted in frequency in line with the changed period of the helix.^{81,82} A region of total reflection of light of any polarization appears in the middle of the region of selective reflection, and here each circularly-polarized component is transformed into itself upon reflection. We should note that the assertion of the circularity of the selectively reflected polarization is approximate, and breaks down at fields close to critical. At fields close to F_c , the region of first-order reflection breaks down into three peaks and the reflection pattern becomes similar to reflection in higher orders (see below). Calculation shows that the splitting of the reflection region occurs at a field of $F \geq 0.98F_c$. For $F \leq 0.98F_c$, the dependence on F of the total widths $\Delta\omega_s$ of the region of selective reflection and $\Delta\omega_T$ of the region of reflection of light of any polarization are given by the expressions

$$\Delta\omega_s = \omega_B \delta (\sqrt{\rho_0^2 + \rho_s^2} + \rho_s), \quad (5.6)$$

$$\Delta\omega_T = \omega_B \delta (\sqrt{\rho_0^2 + \rho_s^2} - \rho_s). \quad (5.7)$$

Curve 1 in Fig. 12 shows the relationship (5.7). The centers of the regions lie at the frequency $\omega_B = 2\pi c/p_F \sqrt{\epsilon}$.

In the first order for $F \geq 0.98F_c$ and in the second and higher orders at all values of F , diffractive reflection is realized in three independent frequency ranges.

In the frequency ranges

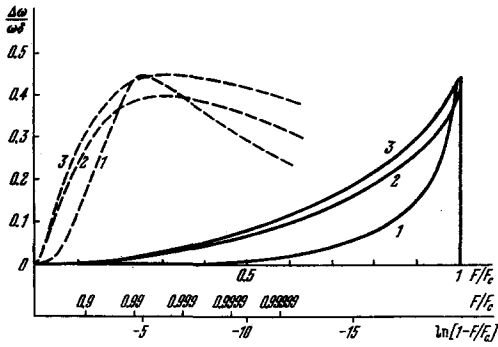


FIG. 12. Field dependence of the frequency intervals of selective reflection with normal incidence of light on a CLC.⁸⁸ Curves 1 and 2 give the width of the regions of total reflection in the first and second orders, respectively, and curve 3 the width of the regions of selective reflection of one polarization in the second order (the dotted curves correspond to the logarithmic scale).

$$-\frac{\delta}{2}(\sqrt{\rho_0^2 + \rho_s^2} + \rho_s) < \frac{\Delta\omega}{\omega} < -\frac{\delta}{2}(\sqrt{\rho_0^2 + \rho_s^2} - \rho_s), \quad (5.8)$$

$$\frac{\delta}{2}(\sqrt{\rho_0^2 + \rho_s^2} - \rho_s) < \frac{\Delta\omega}{\omega} < \frac{\delta}{2}(\sqrt{\rho_0^2 + \rho_s^2} + \rho_s) \quad (5.9)$$

(here $\Delta\omega = \omega - s\omega_B$), only a wave having an elliptical polarization depending on $\Delta\omega$ is reflected. It is determined by the corresponding eigensolution of the system (5.4). For the third and higher orders, the polarizations that undergo selective reflection are close to linear. They are directed along the field for the frequency range (5.8) and perpendicular to the field for the range (5.9). The frequency width of the regions (5.8) and (5.9) is the same, while its relation to F for the second order is shown by curve 3 in Fig. 12.

A wave having any polarization is reflected in the following frequency range that lies between the ranges of (5.8) and (5.9):

$$-\frac{\delta}{2}\left(\frac{\rho_s \sqrt{\rho_0^2 - \rho_s^2}}{\rho_0}\right) < \frac{\Delta\omega}{\omega} < \frac{\delta}{2}\left(\frac{\rho_s \sqrt{\rho_0^2 - \rho_s^2}}{\rho_0}\right). \quad (5.10)$$

The dependence of the width of this region on F for the second order is depicted by curve 2 in Fig. 12. In the third and higher orders in the region (5.10), a wave polarized along the field yields upon reflection a wave polarized perpendicular to the field, and vice versa (see Sec. 4e).

At low fields the frequency ranges (5.8)–(5.10) behave as $(F/F_c)^{2s-2}$. Thus the width of the second-order reflection region is proportional to $(F/F_c)^2$. That is, it is more sensitive to small fields than, e.g., the change in the period of the cholesteric helix, which in small fields is of the order of $(F/F_c)^4$. When $F \geq F_c$ the spatial periodicity of the orientation of the molecules in the CLC vanishes, and there is no diffractive reflection.

We note that, in order that the reflection coefficient in the s th order should be close to unity, i.e., that we can consider the crystal to be thick, the thickness L of the crystal must satisfy the relationship

$$L \gg P_F \left(\frac{\Delta\omega}{\omega}\right)_s^{-1}. \quad (5.11)$$

Here $(\Delta\omega/\omega)_s$ is the frequency range characteristic of the given order s , as determined by Eqs. (5.6)–

(5.10).

We have assumed in deriving the expressions (5.8) and (5.10) that the linear birefringence induced by the field is larger than the circular birefringence in the undistorted CLC,⁸⁷ i.e.,

$$\delta\rho_0 > \frac{\delta_0^2}{8\omega_B(\omega^2 - \omega_B^2)}. \quad (5.12)$$

Evidently in weak enough fields we have $\rho_0 \sim (F/F_c)^2$ and the inequality (5.12) breaks down. Hence, strictly speaking, the performed analysis is valid for strong enough fields that satisfy the condition (5.12).

Second-order reflection of light from a CLC distorted by an external (electric) field has been observed experimentally⁸⁹ in the case of propagation of light along the axis of the helix.

6. ABSORBING CHOLESTERIC LIQUID CRYSTALS

Thus far we have assumed that light is not absorbed in the CLC. Actually such absorption occurs and can be quite significant near the absorption bands of the molecules of the CLC or of specially introduced materials (e.g., dyes). The helical structure of the CLC influences the nature of the light absorption and can alter it radically to the point of almost complete suppression of absorption.

As we know, absorption of light gives rise to an imaginary component in the dielectric permittivity tensor $\hat{\epsilon}$. Hence, in treating the optics of absorbing CLCs, we can use the expressions given above in Secs. 1–4, with the difference that $\hat{\epsilon}_1$, $\hat{\epsilon}_2$, and $\hat{\epsilon}_3$ (the principal values of the tensor $\hat{\epsilon}$) are now complex quantities. For example, if in the studied frequency range in the spectrum of the molecules of the CLC several absorption bands exist that are linearly polarized at angles ψ_j to the long axis of the molecules, then the imaginary components of $\bar{\epsilon} = (\epsilon_1 + \epsilon_2)/2$ and $\epsilon_a = \epsilon_1 - \epsilon_2$ are given by the following expressions^{6,90}:

$$\text{Im } \bar{\epsilon} = \sum_j \eta_j \left[1 + \frac{1}{2} \left(1 - \frac{3}{2} \sin^2 \psi_j \right) S \right], \quad (6.1)$$

$$\text{Im } \epsilon_a = 3S \sum_j \eta_j \left(1 - \frac{3}{2} \sin^2 \psi_j \right).$$

Here the quantity η_j is proportional to the corresponding oscillator strength, and S is the order parameter [cf. (1.2)].

a) Suppression of absorption

First let us examine the effect of absorption on the optical characteristics in the region of selective reflection.^{8,91–93} As usual, the effect of absorption on the reflection of light from a CLC is manifested in a decreased reflection coefficient. However, in view of the fact that reflection in a CLC is diffractive in nature, the absorption of light in the region of selective reflection, and correspondingly the decline in the reflection coefficient, can be substantially reduced. Suppression of absorption under diffraction conditions is well known for x-rays,^{75,76} as well as for Mössbauer γ -quanta⁹⁴ and neutrons,⁹⁵ and is called the Borrmann effect. This problem has been treated for CLCs for the case of normal incidence⁹² and for an arbitrary

angle of incidence of the light.⁹³ It has been shown^{92,93} that absorption can be completely suppressed in a crystal with absorption oscillators directed along the long axis of the molecules (i.e., for $\text{Im}\epsilon_1 \neq 0, \text{Im}\epsilon_2 = \text{Im}\epsilon_3 = 0$) in the case of ideal order of the molecules ($S=1$). This means that the intensity of the wave reflected by a thick crystal at a certain polarization and angle of incidence is equal to the incident intensity (Fig. 13).

For example, in this case with normal incidence of light, complete suppression of absorption and conversion of the reflection coefficient to unity are attained at the frequency $\omega = \omega_B \sqrt{2\epsilon_2 / (\text{Re}\epsilon_1 + \epsilon_2)}$ when circularly polarized light is being diffracted. We can directly convince ourselves of this by using Eqs. (3.5), (3.6), and (3.10).

The physical cause of the decreased absorption consists of the fact that the electric field vector in the wave superposition of (4.12) that arises in the crystal is directed at each point of the CLC perpendicular to the long axes of the molecules, and hence to the absorption oscillators. Such waves do not undergo absorption. Complete suppression is not attained for real crystals, while the peak of the reflection curve (see Fig. 13) does not reach unity, and depends strongly on the order parameter S .

The suppression of absorption is also manifested in circular dichroism. Thus, by using Eqs. (3.13) and (3.15) for thick absorbing CLCs, we can represent the expression for the value of the circular dichroism D in the form

$$D = \text{th} \left[\frac{1}{2} L \text{Im}(\beta_2 - \beta_1) \right]. \quad (6.2)$$

Here β_1 and β_2 are defined by Eq. (3.5).

When $\text{Im}\bar{\epsilon} \gg |\delta|$, the expression (6.2) goes over into

$$D = \text{th} \left[\frac{\kappa^2 L \text{Im}\bar{\epsilon} (\text{Re}^2 \epsilon_a - \text{Im}^2 \epsilon_a) - \gamma \text{Im} \epsilon_a \text{Re} \epsilon_a}{8\tau} \right]. \quad (6.3)$$

Here we have $\gamma = 2[1 - (\omega_B/\omega)^2] \text{Re}\bar{\epsilon}$.

Equation (6.3) implies that the quantity D changes sign as a function of the frequency. The change of sign of D implies that the region of diffractive reflection man-

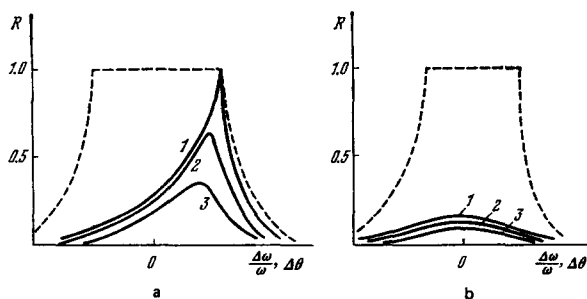


FIG. 13. Qualitative variation of the reflection coefficient R for thick absorbing CLCs at oblique incidence. (a) Reflection of light with the polarization that undergoes suppressed absorption; (b) the polarization that does not undergo suppressed absorption. Curves 1-3 are respectively for values of the order parameter of $S=1, 0.9$, and 0.7 ; dotted curve—in the absence of absorption.

ifests not only suppressed absorption but also an increase in comparison with the mean absorption.

Suppression of light absorption and frequency-dependence of the dichroism have been experimentally observed in Ref. 96 dealing with the change in intensity of a ray transmitted through a CLC in the selective-reflection region.

b) Rotation of the plane of polarization

The existence of anisotropic absorption also strongly alters the frequency dependence of the angle of rotation of the plane of polarization. While a nonabsorbing crystal in the selective reflection region rotates in practically the same manner both to the right and to the left (see Fig. 7), an absorbing crystal rotates preferentially in one direction.^{92,93} This change in the nature of the rotation of the plane of polarization in an absorbing CLC arises from the fact that now the "slow" and the "fast" diffracted waves represented in the expansion (3.7) undergo substantially differing absorptions in the CLC. Consequently one of the waves proves to be suppressed in the CLC, and thus asymmetry arises in the curve of the rotation of the polarization, or else a change in sign of the rotation is completely lacking.

Just as in nonabsorbing CLCs, the rotation angle of the plane of polarization of the light is determined by the phase difference of the terms in the expression (3.15), in which we must now allow for the complex nature of $\bar{\epsilon}$.

Under the same assumptions that we made in deriving (6.3), we find the following expression for the rotation angle of the plane of polarization:

$$\varphi = L \frac{\text{Re}(\beta_1 - \beta_2) - \tau}{2} = \frac{\kappa^2 L}{8\tau} \frac{\gamma (\text{Re}^2 \epsilon_a - \text{Im}^2 \epsilon_a) + 4 \text{Im}\bar{\epsilon} \text{Im} \epsilon_a \text{Re} \epsilon_a}{\gamma^2 + 4 \text{Im}^2 \bar{\epsilon}}. \quad (6.4)$$

Figure 14 shows the form of the relationships described by Eq. (6.4).

Now let us study the propagation of light along the axis of the helix for frequencies ω outside the region of selective reflection. It turns out that here circular dichroism and optical rotatory dispersion are observed at frequencies corresponding to the absorption bands of the molecules.^{90,91,97-103} These are related to the helical structure of the CLC, and they are manifested even in the absence of intrinsic optical activity of the molecules. The observed circular dichroism and

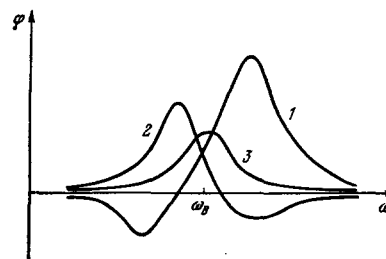


FIG. 14. Qualitative frequency dependence of the angle of rotation of the plane of polarization for different values of absorption in a CLC. 1— $\text{Re} \epsilon_a > \text{Im} \epsilon_a$; 2— $\text{Re} \epsilon_a < \text{Im} \epsilon_a$; 3— $\text{Re} \epsilon_a = \text{Im} \epsilon_a$.

optical rotatory dispersion are determined by the characteristics (in particular, the polarization) of the absorption bands (which enter into the theory in terms of $\hat{\epsilon}$). Outside the region of selective absorption, they are described by Eqs. (6.2) and (6.4) for any thickness L of the crystal and for an arbitrary value of $\text{Im}\hat{\epsilon}$.

A detailed treatment of the optical activity and dichroism of CLCs outside the region of selective reflection can be found in Ref. 91. In this case the validity of Eqs. (6.2)–(6.4) has been tested experimentally in Ref. 90. Here it was shown that the circular dichroism spectra of CLCs actually are determined by the linear dichroism spectrum of the molecules forming the CLC. We should note, as we see from (6.3), that the circular dichroism of CLCs occurs, in particular, even with isotropic absorption of the molecules ($\text{Im}\epsilon_a = 0, \text{Im}\bar{\epsilon} \neq 0$). Thus, one can draw conclusions regarding the structure of the molecules and of the CLC from the measured circular-dichroism spectra.

In Refs. 48 and 49, the circular dichroism and the optical rotatory dispersion of CLCs have been measured near the long-wavelength edge of an electronic absorption band. However, a complete theoretical interpretation of these results has not yet been obtained.

7. IMPERFECT CHOLESTERIC LIQUID CRYSTALS

The ideal cholesteric structure shown in Fig. 2 is rather hard to obtain in thick specimens. The so-called focal-conic configuration, 1, 2, 5 in which the local orientation of the optic axis of the CLC differs at different points, is usually produced in the isotropic liquid-CLC transition, and the crystal scatters light in all directions. Hence one is often dealing with nonideal CLCs in experimental situations.

The plane configuration (up to thicknesses of 100–200 μm) is obtained by applying external fields to a CLC^{13,80} or by mechanical action during the preparation of the specimens.^{1,2,16,18} One can also orient the axis of a CLC parallel to the surface in thin films by coating the plates with surface-active substances.

As we shall see, the imperfect structure of a CLC, and consequently, the lack of coherence in light scattered by individual crystallites of the CLC, is manifested in the intensity and polarization characteristics of the scattering. The fundamental manifestation of imperfection of a CLC in the polarization properties of scattering proves to be depolarization of the scattered light (for a perfect structure the scattered light is completely polarized). Imperfect crystals also show a dependence on the thickness of the specimen differing from ideal ones of the polarization characteristics of the light scattered and transmitted by the CLC.

The physical cause of the stated differences is quite understandable. The relationship of the polarization of the light to the thickness stems from the birefringence of the CLC. The birefringence causes the polarization of the direct wave to vary as it penetrates into the interior of the specimen. Analogous changes occur in the

polarization of the scattered wave as it propagates in the specimen. Consequently the polarization of the scattered wave emerging from the specimen proves to depend on the point at which the scattering event occurred, while on the whole the polarization characteristics show a dependence on the thickness of the specimen. Depolarization arises from the fact that the observed scattered radiation amounts to an incoherent superposition of the waves scattered from the individual regions of the CLC.

a) Thin imperfect crystals

We can apply the kinematical theory to describe light scattering from thin imperfect specimens having a scatter of orientation of the optic axis of the individual crystallites. On the basis of the Bragg condition and the laws of refraction of light at the boundaries of a CLC, Ferguson¹⁵ and subsequently other authors^{104,105} have derived formulas for this case that relate the angles of incidence and reflection to the wavelength corresponding to the maximum selective reflection of light for the cases depicted in Figs. 15a and b respectively.

$$\lambda = np \cos \left[\frac{1}{2} \arcsin \left(\frac{\sin \varphi_i}{n} \right) + \frac{1}{2} \arcsin \left(\frac{\sin \varphi_r}{n} \right) \right], \quad (7.1a)$$

$$\lambda = np \sin \left[\frac{1}{2} \arcsin \left(\frac{\sin \varphi_i}{n} \right) + \frac{1}{2} \arcsin \left(\frac{\sin \varphi_r}{n} \right) \right]. \quad (7.1b)$$

Here λ is the wavelength of the light *in vacuo*, and $n = \sqrt{\bar{\epsilon}}$ is the refractive index of the CLC. The more general case in which the diffracted ray does not lie in the plane formed by the incident ray and the normal to the surface of the CLC has been treated in Ref. 106.

The formulas (7.1) are purely geometrical and they tell nothing about the intensity and polarization of the diffracted beam. As has been shown,¹⁰⁴ study of the dependence of the diffraction intensity on the angle φ_r for fixed φ_i (see Fig. 15) and on the wavelength allow one to draw conclusions concerning the amount of disorientation of the crystallites in the crystal. In all cases the measurements show that the strongest reflection occurs at $\varphi_r = \varphi_i$. That is, the specimens consist of crystallites whose axes are mainly parallel to the surface in the case of a focal-conic configuration and perpendicular in the case of a plane configuration.^{104–106}

Now let us examine in greater detail the polarization properties of diffraction by imperfect crystals. We shall assume below for the sake of concreteness that the imperfection of the CLC is of a type analogous to a mosaic single crystal, rather than a polycrystal. This

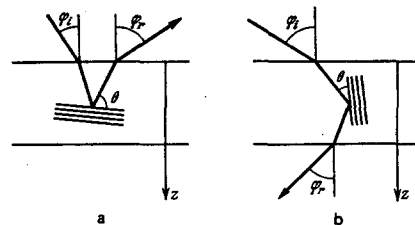


FIG. 15. Possible scattering geometries in imperfect CLC specimens.

implies that the individual regions of the CLC scatter incoherently, though their spatial orientation differs little. Under these conditions, in line with the incoherence of the interference in the scattering from the individual regions of the CLC, it is convenient to transform from the equations for the amplitudes of the fields to equations for quantities quadratic in the amplitudes. Below we shall employ as such the polarization tensors of the direct and the diffracted waves. The polarization tensor \hat{I} (see, e.g., Ref. 107) differs only in normalization from the polarization density matrix:¹⁰⁸ its components are quadratic in the amplitude of the field, while $\text{Sp} \hat{I}$ gives the intensity of the wave.

At first we shall consider that the specimen is thin enough that we can neglect the attenuation of the wave in the specimen and that it suffices to take into account only single diffractive scattering. Let a wave be incident on the crystal that is characterized by the polarization tensor \hat{I}^i . When light propagates at an angle to the optic axis, its polarization is altered by the birefringence. The variation of the polarization tensor \hat{I}_0 of the incident wave is described by the equation

$$\frac{d\hat{I}_0}{dz} = \hat{A}^0 \hat{I}_0. \quad (7.2)$$

Here we have the tensor $A_{iklm}^0 = i(\omega/c)[(\hat{\epsilon}_0)_{ik}^0 \delta_{lm} - \delta_{ik}(\hat{\epsilon}_0)_{lm}^0] / \cos \varphi_i'$, $i, k, l, m = 1, 2$, while the components of the tensor $\hat{\epsilon}_0$ from (2.2) employed here describe the propagation of light in the direction \mathbf{k}_0 . That is, they are written in a system of coordinates, two of whose axes (1 and 2) are orthogonal to the direction of propagation of light, while the third coincides with this direction. Also we have $\cos \varphi_i' = k_{0z} / |\mathbf{k}_0|$.

We get the following expression from (7.2) for the polarization tensor \hat{I}_0 at depth z from the surface of the specimen:

$$\hat{I}_0(z) = \exp(\hat{A}^0 z) \hat{I}_0. \quad (7.3)$$

The polarization tensor \hat{I}_1 of the scattered wave is formed by the diffraction of the incident wave by the crystallites lying at different depths. If light undergoes Bragg scattering at depth z from the wave \mathbf{k}_0 to the wave \mathbf{k}_1 , then the increment $\Delta \hat{I}_1$ to \hat{I}_1 arising from this scattering is described by the expression

$$(\Delta \hat{I}_1)_{ik} = C_{iklm}^0 (\hat{I}_0(z))_{lm} \Delta z. \quad (7.4)$$

Here Δz is the characteristic dimension of the crystallite, we have $C_{iklm}^0 = r F_{i1} F_{k2}^*$, r is a coefficient that depends on the mean dimensions and shapes of the crystallites, and the tensor \hat{F} coincides with the structure amplitude of (2.5). That is, its components are determined by the relationship

$$F_{ij} = F(\mathbf{k}_0, \mathbf{e}_{0i}; \mathbf{k}_1, \mathbf{e}_{1j}) = e_{1i}^* \hat{\epsilon}_{ij} e_{0j}. \quad (7.5)$$

In order to obtain the polarization tensor \hat{I}^r of the wave scattered by the entire specimen, we must integrate Eq. (7.4) over the thickness of the specimen, taking into account the fact that the polarization tensor $\Delta \hat{I}_1$ varies with the coordinate z as light propagates from the depth z to the surface of the crystal according to a law analogous to (7.2) and (7.3). Consequently the expression for \hat{I}^r acquires the form

$$\hat{I}^r = \int_0^L \exp(z \hat{A}^1) \hat{C}^{1*} \exp(z \hat{A}^0) dz \hat{I}^i. \quad (7.6)$$

Here the tensor \hat{A}^1 is determined by the expression for \hat{A}^0 [see (7.2)] by employing in it the components of the tensor $\hat{\epsilon}_0$ that describe the propagation of light in the scattering direction. That is, they are written in a system of coordinates, two axes of which (1 and 2) are perpendicular to the direction of propagation of the scattered light, while the third axis coincides with this direction.

The polarization tensor (7.6) determines all the characteristics of the diffracted beam, including its polarization characteristics.^{107, 108}

Equation (7.6) implies that the polarization characteristics of scattering by a mosaic crystal differ from the case of an ideal specimen. In particular, the scattered light is depolarized (even with a fully polarized incident beam, the scattered light proves partially polarized).

In the general case the expression (7.6) for \hat{I}^r proves rather unwieldy after integration. If the incident light is unpolarized (i.e., $(\hat{I}^i)_{ik} \sim \delta_{ik}$), then formula (7.6) is simplified, and yields the following expression for the polarized density matrix of the diffracted beam $\hat{\rho}^r = \hat{I}^r / \text{Sp} \hat{I}^r$:

$$\rho_{11}^r = \frac{1}{1 + \sin^2 \theta}, \quad \rho_{22}^r = \frac{\sin^2 \theta}{1 + \sin^2 \theta}, \quad \rho_{12}^r = \rho_{21}^r = \frac{\sin \theta \cos \varphi_r' \{ \exp[-2i\kappa \delta L \sin^2 \varphi_r' / \cos \varphi_r'] - 1 \}}{2\kappa \delta L \sin \varphi_r' (1 + \sin^2 \theta)}. \quad (7.7)$$

Here we have $\cos \varphi_r' = k_{1z} / |\mathbf{k}_1|$.

We see from (7.7) that we can neglect the nondiagonal elements in (7.7) for a thick enough crystal (when $L \gg \sin \theta \cos \varphi_r' / \kappa \delta \sin \varphi_r'$). Then the light emerging from the crystal proves to be partially linearly polarized along σ with the degree of polarization $P = \cos^2 \theta / (1 + \sin^2 \theta)$. Thus, for small θ the light proves to be almost fully polarized perpendicular to the axis of the helix. We note that Eq. (7.7) also holds in the case shown in Fig. 15b.

In the above discussion we have assumed that the light undergoes only single Bragg scattering inside the specimen (an analogous approach has been employed for describing diffraction of Mössbauer radiation in mosaic crystals^{109, 110}).

b) Multiple scattering taken into account

For thick CLC specimens, multiple scattering from the incident to the reflected wave and vice versa can become substantial. This involves the fact that the intensity of the diffracted wave in a thick specimen is not small in comparison with that of the incident wave, and we must allow for the fact that it also undergoes diffractive scattering. We can account for the diffractive scattering of this wave (as well as the birefringence in the CLC) in the same way as we did above. However here we must bear in mind the attenuation of the waves in the specimen owing to diffractive scattering.¹¹¹

Here it turns out that the variation with the coordinate in the specimen of the polarization tensors \hat{I}_0 and \hat{I}_1 of

the primary and diffracted waves is described by the equations

$$\begin{aligned}\frac{d\hat{I}_0}{dz} &= \hat{A}^0 \hat{I}_0 - \hat{B}^0 \hat{I}_0 + \hat{C}^{01} \hat{I}_1, \\ \frac{d\hat{I}_1}{dz} &= \hat{A}^1 \hat{I}_1 - \hat{B}^1 \hat{I}_1 + \hat{C}^{10} \hat{I}_0.\end{aligned}\quad (7.8)$$

The tensors \hat{A}^0 and \hat{A}^1 determine the variations in \hat{I}_0 and \hat{I}_1 with the coordinate z arising from birefringence, and they have been introduced above in (7.2). The tensors \hat{B}^0 , \hat{B}^1 and \hat{C}^{01} , \hat{C}^{10} determine the variation in \hat{I}_0 and \hat{I}_1 associated with Bragg scattering (\hat{B}^0 and \hat{B}^1 owing to loss, and \hat{C}^{01} and \hat{C}^{10} owing to gain of radiation by the primary or the diffracted wave). They are expressed in terms of $\hat{\epsilon}_{\pm 1}$ [the structure amplitude of (2.5)].¹¹¹ For example, the expression for \hat{C}^{10} is defined in (7.4), while $C_{iklm}^{01} = C_{imlk}^{10}$. Here \hat{B}^0 , \hat{B}^1 , \hat{C}^{01} , and \hat{C}^{10} prove to be functions of the frequency of the light incident on the crystal and they differ from zero only when the Bragg condition (2.4) is satisfied. We note that the equations (7.8) are analogous to the transport equations employed in describing radiation in turbid media¹¹² and in a magnetoactive plasma.^{107, 113} The difference consists of the fact that only Bragg scattering and zero-angle scattering prove substantial in the CLC case.

Similar equations are used to describe the intensities of the direct and the diffracted waves in studying diffraction of x-rays^{114a} and neutrons^{114b} by mosaic crystals. In a CLC in the general case the equations for the intensities (diagonal elements of the tensors \hat{I}_0 and \hat{I}_1) and for the nondiagonal elements of \hat{I}_0 and \hat{I}_1 cannot be separated owing to the complex polarization properties of the scattering and owing to birefringence, which complicates the solution of the problem as compared with the x-ray case.

c) Normal incidence of light

We give below the solutions of the system (7.8) in the simplest case with normal incidence of the light on the specimen.¹¹¹ In this case the equations (7.8) as written in the circular unit vectors n_+ and n_- separate into several uncoupled systems of equations. With the boundary conditions $\hat{I}_0(z=0) = \hat{I}^i$, $\hat{I}_1(z=L) = 0$ taken into account their solution has the following form for the reflected wave:

$$\begin{aligned}I_{11}^i &= I_{11}^i \frac{\mu_d \operatorname{sh}(\tilde{\mu}L)}{(\mu + \mu_d) \operatorname{sh}(\tilde{\mu}L) + \tilde{\mu} \operatorname{ch}(\tilde{\mu}L)}, \\ I_{12}^i &= I_{12}^i = I_{21}^i = 0, \quad \tilde{\mu} = \sqrt{\mu^2 + 2\mu\mu_d}.\end{aligned}\quad (7.9)$$

Here the indexes 1 and 2 refer respectively to the polarizations n_+ and n_- , μ is the linear absorption coefficient, and μ_d^{-1} is the diffractive extinction length. The quantity μ_d depends sharply on the frequency, reaching a maximum when $\omega = \omega_B$, and vanishing outside the Bragg condition. The form of the frequency dependence of μ_d is determined by the dimensions of the crystallites and by their angular disorientation in the specimen.¹¹¹

The expressions (7.9) yield a quite natural result: only light having the polarization n_+ undergoes reflection out of the entire incident beam, and the reflected light has the same polarization. The reflection coef-

ficient for this polarization is simply the coefficient of I_{11}^i in (7.9). If the absorption is small, so that $\tilde{\mu}L \ll 1$, then we have

$$I_{11}^i = \frac{\mu_d L}{1 + \mu_d L} I_{11}^i. \quad (7.10)$$

Hence we see that the reflection coefficient has an appreciable value when $\mu_d L \gtrsim 1$. Here the frequency width of the curve for reflection from a thick nonideal crystal is substantially elevated as compared with that of the function μ_d .

For the transmitted wave we have

$$I_{11}^t = \frac{\tilde{\mu} I_{11}^i}{(\mu + \mu_d) \operatorname{sh}(\tilde{\mu}L) + \tilde{\mu} \operatorname{ch}(\tilde{\mu}L)}, \quad (7.11)$$

$$I_{22}^t = I_{22}^i e^{-\mu L}, \quad (7.12)$$

$$I_{12}^t = I_{21}^t = I_{11}^i \exp\left[-\left(\mu + \frac{\mu_d}{2} + 2i\mu_r\right)L\right]. \quad (7.13)$$

Here μ_r is the mean rotatory power of the plane of polarization associated with diffractive scattering, as determined by the properties of the individual crystallites of the specimen.¹¹¹

We see from (7.11) and (7.12) that the intensity of the transmitted wave of polarization n_+ is diminished by diffractive reflection more rapidly with increasing thickness than that of the wave of polarization n_- , which undergoes only the ordinary attenuation.

Another qualitative feature is that the transmitted light is generally depolarized. As we can see from (7.11)–(7.13), the degree of polarization of the transmitted light is minimal for a quite definite polarization of the incident light (this polarization depends on the thickness of the specimen and is determined by the condition $I_{11}^t = I_{22}^t$). In this case the emerging light is partially linearly polarized and its degree of polarization is:

$$P_{\min} = \sqrt{\left| \operatorname{ch}(\tilde{\mu}L) + \frac{\mu + \mu_d}{\tilde{\mu}} \operatorname{sh}(\tilde{\mu}L) \right| \exp[-(\mu + \mu_d)L]}. \quad (7.14)$$

However, light having the circular polarizations n_+ and n_- is not depolarized.

d) The case of linearly polarized light

Let us study as an example the case of linearly polarized incident light, for which the polarization tensor has the form

$$\hat{I}^i = \frac{I^i}{2} \begin{pmatrix} 1 & -i \\ i & 1 \end{pmatrix}. \quad (7.15)$$

Here I^i is the intensity of the incident light (we recall that we are using circular polarization vectors as the basis).

As we have noted, the reflected light proves to be circularly polarized, while the light transmitted through the specimen is generally partially polarized (elliptically).

The angle of rotation φ of the polarization ellipse on emergence is given by the expression

$$\operatorname{tg} 2\varphi = \frac{\operatorname{Re} I_{12}^t}{\operatorname{Im} I_{12}^t} = \operatorname{tg}(2\mu_r L). \quad (7.16)$$

We see that, in contrast to ideal CLCs [see (3.16)], φ is proportional to the thickness of the crystal.

If the absorption coefficient μ is small, so that

$\mu_d L \ll 1$, then we obtain from (7.11)–(7.13) simple expressions also for the rest of the polarization characteristics:

for the degree of polarization we have:

$$P = \sqrt{\mu_d^2 L^2 + 4(1 + \mu_d L)^2 \exp(-\mu_d L)(2 + \mu_d L)^{-1}}, \quad (7.17)$$

for the ratio of axes of the polarization ellipse:

$$b = \mu_d L \left\{ 2(1 + \mu_d L) \exp(-\mu_d L/2) + \sqrt{\mu_d^2 L^2 + 4(1 + \mu_d L)^2 \exp(-\mu_d L)} \right\}^{-1}, \quad (7.18)$$

and for the intensity of the transmitted light:

$$I^t = I^i \frac{1 + (\mu_d L/2)}{1 + \mu_d L}. \quad (7.19)$$

Figure 16 shows the thickness dependence of P , b , and I^t : at small thickness we have $P = 1$ and $b = 0$, since the incident light is linearly polarized, and the effect of the crystal is small. With increasing L , the degree of polarization first falls (owing to multiple incoherent scattering) and the transmitted beam becomes partially polarized. However, with further increase in thickness the degree of polarization again approaches unity, since only the polarization n_{\perp} is transmitted through a very thick crystal. We recall that polarized light does not undergo any depolarization at all in passing through an ideal crystal.

We see from studying the case of normal incidence that the circular polarizations n_{\perp} and n_{\parallel} propagate independently in the CLC, with only the polarization n_{\perp} being reflected. However, nonideal CLCs possess a nondiffractive "pumping" mechanism of the polarization n_{\perp} into n_{\parallel} (and vice versa). As a result this leads also to selective attenuation of the light of polarization n_{\perp} . A source of this pumping is, e.g., optical inhomogeneities of a mosaic crystal.¹¹¹ We recall that selective attenuation of the light having the undiffracted polarization has been observed by Kizel' and his associates.^{48, 49}

We note in closing this section that the optical properties of imperfect CLCs have not yet been sufficiently studied quantitatively. In addition to the mosaic CLCs discussed above, Refs. 86, 115 have investigated CLCs having a gradient of the pitch of the helix.

8. CHIRAL SMECTIC LIQUID CRYSTALS

The theory presented above of the optical properties of CLCs can be applied to chiral smectic liquid crystals (SLC*), interest in which has recently heightened in connection with the detection in them of ferroelectric

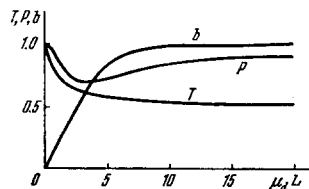


FIG. 16. Dependence of the transmission coefficient $T = I^t/I^i$, the degree of polarization P , and the ratio of axes of the polarization ellipse b in the transmitted beam on the thickness of a mosaic specimen for linearly polarized incident light.¹¹¹

properties.^{5, 116} Here it turns out that on the whole their optical properties are analogous to those of CLCs. Yet there are certain qualitative differences caused by the difference in structures of these varieties of liquid crystals.

Several varieties of SLC*'s are known.^{5, 117} Just like other smectic crystals, SLC*'s are layered structures consisting of monomolecular layers with the interaction between the layers being weaker than the interaction of molecules within a layer.¹¹⁷⁻¹²³ The structure of a SLC* can be represented schematically by Fig. 1, which depicts the structure of a CLC, by raising the long axes of the molecules therein at the same given angle out of the planes without at the same time altering their azimuthal angle.

a) Dielectric properties of chiral smectic liquid crystals

The differences in structure of CLCs and SLC*'s give rise to a difference in the z -dependence of their dielectric-permittivity tensors. While in a CLC the orientation of only two principal axes of the tensor $\hat{\epsilon}(z)$ varies with z (the direction of the third axis coincides with z), in a SLC* the orientation of all three principal axes of the tensor $\hat{\epsilon}(z)$ vary with varying z . That is, all its components vary. In agreement with the structure, the periodicity of the variation of the dielectric properties of a SLC* along z coincides with the pitch p of the helix (in a CLC the corresponding period is $p/2$).

Naturally the optical properties of a SLC* are determined by the form of the tensor $\hat{\epsilon}(z)$. Without writing out the explicit form of $\hat{\epsilon}(z)$ in the r -representation, we shall give its Fourier expansion that is employed below.

For a SLC* the components $\hat{\epsilon}_s$ in the Fourier expansion (2.2) are defined by the following relationships:

$$\left. \begin{aligned} \hat{\epsilon}_1 = \hat{\epsilon}_2 = \frac{1}{4}(\epsilon_1 - \epsilon_2) \sin 2\theta \begin{pmatrix} 0 & 0 & \pm i \\ 0 & 0 & 1 \\ \pm i & 1 & 0 \end{pmatrix}, \\ \hat{\epsilon}_3 = \hat{\epsilon}_4 = \frac{1}{4}(\epsilon_1 - \epsilon_2 \cos^2 \theta - \epsilon_3 \sin^2 \theta) \begin{pmatrix} 1 & \mp i & 0 \\ \mp i & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \\ \hat{\epsilon}_s = 0 \text{ when } |s| \geq 3. \end{aligned} \right\} \quad (8.1)$$

The upper sign in (8.1) corresponds to a right-hand and the lower to a left-hand helix.

Only the diagonal components of the tensor $\hat{\epsilon}_0$ differ from zero:

$$\left. \begin{aligned} (\hat{\epsilon}_0)_{11} = (\hat{\epsilon}_0)_{22} = \frac{1}{2}(\epsilon_1 + \epsilon_2 \cos^2 \theta + \epsilon_3 \sin^2 \theta), \\ (\hat{\epsilon}_0)_{33} = \epsilon_2 \sin^2 \theta + \epsilon_3 \cos^2 \theta. \end{aligned} \right\} \quad (8.2)$$

Here ϵ_1 , ϵ_2 , and ϵ_3 are the principal values of the dielectric permittivity tensor, while θ is the angle that the principal axis of the tensor $\hat{\epsilon}(z)$ corresponding to ϵ_3 makes with the z axis (we assume that the axis corresponding to ϵ_2 lies in the plane of the smectic layer). Let us call attention to the fact that, in line with the noted difference in the periodicity of the properties of SLC*'s and CLCs, we should not assume $\tau = 2\pi/p$ in the expansion (2.2) (rather than $4\pi/p$, as in a CLC). We note that the Fourier expansion of (8.1) does not take into account the periodicity of the SLC* associated

with its layered structure. In view of the smallness of the corresponding period, the latter can be manifested only in x-ray scattering.

b) Second order diffractive reflection

We see from (8.1) that the second harmonic in the Fourier expansion of $\hat{\varepsilon}(z)$ for a SLC* coincides, apart from a coefficient, with the first harmonic in the corresponding expansion (2.2) for a CLC. This means that the second-order diffractive reflection in a SLC* is qualitatively analogous to the first order in a CLC. A quantitative analysis can be conducted by analogy with the case of a CLC employing in the equations (4.4) the Fourier components $\hat{\varepsilon}_0$ and $\hat{\varepsilon}_{\pm 2}$ from Eqs. (8.1) and (8.2).

As analysis of the exact solution known for this case shows, only the second order of diffractive reflection is realized when light propagates along the optic axis of a SLC*. In this case the optical properties of the SLC* prove fully analogous to the properties of a CLC, and they are described by the expression in Sec. 3, with the substitution^{11a}:

$$\bar{\varepsilon} \text{ by } \bar{\varepsilon}' = \frac{\varepsilon_1 + \varepsilon_2 \cos^2 \theta + \varepsilon_3 \sin^2 \theta}{2} - \frac{(\varepsilon_2 - \varepsilon_3)^2 \sin^2 2\theta}{8\varepsilon_3},$$

$$\delta \text{ by } \delta' = \left[\frac{\varepsilon_1 - \varepsilon_2 \cos^2 \theta - \varepsilon_3 \sin^2 \theta}{2} + \frac{(\varepsilon_2 - \varepsilon_3)^2 \sin^2 2\theta}{8\varepsilon_3} \right] \frac{1}{\bar{\varepsilon}'}$$

c) First order reflection

Thus, qualitative differences in the optical properties of SLC*'s as compared with CLCs can be manifested (and as we shall see, they are manifested) only for light propagating at an angle to the optic axis, and only in the first order reflection. Hence we shall restrict the treatment below of the optical properties of SLC*'s to the first order of diffractive reflection. In this case, when we substitute $\hat{\varepsilon}$ in the form (8.1) and (8.2) into Eq. (4.1), the system (4.4) of equations breaks down into two independent systems:

$$\begin{cases} \left(\varepsilon_\sigma - \frac{k_z^2}{\kappa_0^2} \right) E_0^\sigma + l E_1^\pi = 0, & \left(\varepsilon_\pi - \frac{k_z^2}{\kappa_0^2} \right) E_0^\pi + l E_1^\sigma = 0, \\ l E_0^\sigma + \left(\varepsilon_\pi - \frac{k_z^2}{\kappa_0^2} \right) E_1^\pi = 0, & l E_0^\pi + \left(\varepsilon_\sigma - \frac{k_z^2}{\kappa_0^2} \right) E_1^\sigma = 0. \end{cases} \quad (8.3)$$

Here E^σ and E^π are the amplitudes of the σ - and π -polarized waves, and we have $\kappa_0^2 = \omega^2/c^2$, $\varepsilon_\sigma = (\hat{\varepsilon}_0)_{11}$, $\varepsilon_\pi = (\hat{\varepsilon}_0)_{33} \sin^2 \theta + (\hat{\varepsilon}_0)_{33} \cos^2 \theta$, $l = (1/4)(\varepsilon_2 - \varepsilon_3) \sin 2\theta \cos \theta$, and $(\pi/2) - \theta$ is the angle between the optic axis and the direction of propagation of the light.

Equation (8.3) implies that a wave with σ -polarization yields a π -polarized wave upon diffraction and vice versa.

The solution of each of the systems of (8.3) and of the corresponding boundary problem is simpler than in the case of a CLC, and it is fully analogous to the solutions in the case of x-ray diffraction (see, e.g., Refs. 75-77). Hence we shall directly present the final results, as above, for a SLC* specimen in the form of a plane-parallel plate.

d) The boundary problem

Whenever the axis of the helix is perpendicular to the surface of the SLC* (the Bragg case), the angular (fre-

quency) reflection regions for σ - and π -polarizations given by the systems (8.3) coincide. Here the polarization and frequency (angular) dependences of the reflection coefficient are described by the expression

$$R = |e_1^* \hat{\varepsilon}_1 e_0|^2 \frac{\sin^2 (\kappa_0 L \sqrt{\Delta_B^2 - l^2/2} \sqrt{\bar{\varepsilon}' \sin \theta})}{\Delta_B^2 - l^2 \cos^2 (\kappa_0 L \sqrt{\Delta_B^2 - l^2/2} \sqrt{\bar{\varepsilon}' \sin \theta})}. \quad (8.4)$$

Here e_0 and e_1 are the polarization vectors of the direct and the diffracted waves, and we have $\bar{\varepsilon}' = (\varepsilon_\sigma + \varepsilon_\pi)/2$. L is the thickness of the crystal, and the parameter Δ_B , as before, characterizes the deviation from the Bragg condition: $\Delta_B = (\tau/2\kappa_0^2)(2\kappa_0 \sqrt{\bar{\varepsilon}' - \cos^2 \theta} - \tau)$. We note that the result of summing Eq. (8.4) over the final polarizations e_1 does not depend on e_0 , i.e., only the polarization of the scattered wave depends on the polarizations of the primary wave, but not its intensity. The middle of the reflection region lies at $\Delta_B = 0$, while the boundaries are determined by the relationship $\Delta_B = \pm 1$.

One can easily reveal the polarization characteristics of the scattering by analyzing the factor $|e_1^* \hat{\varepsilon}_1 e_0|^2$ in (8.4). Thus it turns out that in the first order of diffractive reflection from a SLC*, a right-circularly polarized wave converts into a left-hand one and vice versa (we recall that in normal incidence in a CLC, one circular polarization is not reflected at all, while the wave that interacts with the CLC retains its polarization upon scattering). In scattering of linearly polarized light, the scattered wave is polarized linearly, while the angles φ_0 and φ_1 formed by the plane of polarization with the plane of scattering in the incident and scattered waves, respectively, are coupled by the relationship $\varphi_1 = (\pi/2) - \varphi_0$.

Whenever the axis of the helix is parallel to the plane of the specimen (the Laue case), the regions of reflections for σ - and π -polarizations generally do not coincide, though they can partially overlap. Here, if we assume that $L \gg \kappa_0 p^2 \cos \theta$ (i.e., Bragg diffraction rather than Raman-Nath^{14b} diffraction is realized), we find that the reflection coefficient depends on the polarization of the incident wave, while its frequency-(angular) dependence for σ - and π -polarizations is given by the expression

$$R = \frac{l^2}{(\Delta_L \pm m)^2 + l^2} \sin^2 \left(\frac{\kappa_0 L \sqrt{(\Delta_L \pm m)^2 + l^2}}{2 \sqrt{\bar{\varepsilon}' \cos \theta}} \right). \quad (8.5)$$

Here we have

$$m = \frac{\varepsilon_\sigma - \varepsilon_\pi}{2} = \frac{\cos^2 \theta}{4} |\varepsilon_1 + \varepsilon_2 (1 - 2 \sin^2 \theta) + \varepsilon_3 (1 - 3 \cos^2 \theta)|,$$

$$\Delta_L = \frac{\tau}{2\kappa_0^2} (\tau - 2\kappa_0 \sin \theta).$$

The plus sign in (8.5) gives the reflection coefficient for σ -polarized light, and the minus sign for π -polarized light. We note that when $m = 0$ the reflection zones coincide, and the polarization properties in this case are the same as in the Bragg case. As Eq. (8.5) implies, in the Laue case the reflection coefficient R depends periodically on the thickness L . This is the so-called pendulum beating that is well known in x-ray diffraction.⁷⁵⁻⁷⁷

The analytical results given above (exact and obtained within the framework of the dynamical theory

of diffraction) agree with the results of numerical calculations of the optical properties of SLC*⁷s.¹²² We note also that we have employed for simplicity a restriction on the form of the tensor $\hat{\epsilon}(z)$ for the SLC* that is justified by physical considerations (one of the principal axes of $\hat{\epsilon}(z)$ lies in the plane of the smectic layer). Removal of this restriction¹²² yields no qualitatively new effects and introduces no fundamental complications in the solution of the problem. However, we do not present the appropriate expressions here owing to their unwieldiness.

We should note that the same orientation of the axes of the molecules with respect to the optic axis as in a SLC* (and hence analogous optical properties) can be realized in a CLC near phase-transition points¹²⁴ or in a field parallel to the axis of the helix.⁸² Thus, e.g., in a CLC placed in an external field, diffraction of light corresponding to a periodicity of p (rather than $p/2$) has been observed.¹²⁵ However, it has not been ruled out that the periodicity observed in Ref. 125 involves a domain structure of the specimen (see, e.g., Ref. 126).

9. COHERENT RADIATION FROM FAST CHARGED PARTICLES IN CHOLESTERIC LIQUID CRYSTALS

Above we have discussed the optical properties of CLCs. Naturally, the complicated spatial structure of CLCs and the consequent anisotropic and spatially inhomogeneous dielectric properties of CLCs must also be manifested in the coherent radiation from fast charged particles. A number of theoretical studies^{127-130,147} have been concerned with this problem, in which interesting qualitative features have been revealed in Vavilov-Čerenkov radiation in CLCs. It has turned out that in a CLC, as compared with a homogeneous medium, the angular distribution of the Vavilov-Čerenkov radiation varies substantially (two cones of Čerenkov radiation are realized). Moreover, there exists a coherent radiation that is absent in homogeneous media, the so-called structural Vavilov-Čerenkov radiation.¹²⁸ This radiation arises from the spatial periodicity of the CLC, and in contrast to Vavilov-Čerenkov radiation, it exists both at a particle velocity v above the phase velocity of light $c_{ph} = c/\sqrt{\bar{\epsilon}}$ and when $v < c_{ph}$.

We present below the problems of the theory of coherent radiation from charged particles in a CLC.

a) Kinematical treatment

The radiation that accompanies a uniformly moving particle in a medium is caused by the coherent radiation from the atoms of this medium polarized by the field of the particle. An extensive literature has been devoted to presenting this problem for media of simple structure (see, e.g., Refs. 79, 131, 132). In a medium of complicated structure such as a CLC, this radiation has a number of peculiarities. We shall present its fundamental characteristics on the basis of a kinematical treatment. When a particle is moving in a periodic medium, the condition for coherent superposition of the radiation from the individual atoms leads to the follow-

ing relation between the direction of emission and its frequency ω :

$$\omega \left(1 - \frac{v}{c_{ph}} \cos \theta_e \right) = s\tau v \cos \psi. \quad (9.1)$$

Here θ_e is the angle between the velocity of the particle and the direction of emission, ψ is the angle between the velocity of the particle and the axis of the cholesteric helix, $\tau = 4\pi/p$, and s is an integer. When $s = 0$ we get the well-known condition for Čerenkov radiation $\cos \theta_e = c_{ph}/v$. When $s \neq 0$, radiation exists only in spatially periodic media, and is called structural Vavilov-Čerenkov radiation (in the literature this radiation is also called quasi-Čerenkov, resonance, and transition radiation in a periodic medium). Below we shall take up in greater detail the structural radiation in a CLC.

The relationship between the direction and frequency for a CLC that is given by Eq. (9.1) is fully analogous to the case of radiation in periodic media having a scalar dielectric permittivity.^{132,133} However, in CLCs, in line with the form of the tensor $\hat{\epsilon}(z)$ (2.2) for structural radiation s in Eq. (9.1) can adopt only values of ± 1 . The fundamental features of structural radiation in a CLC are manifested in its polarization properties.

For the sake of definiteness, we shall treat below the motion of a particle along the optic axis of a planar specimen of a CLC of thickness L . The field E_1 of the structural radiation is determined by solving the equation

$$\Delta E_1 - \frac{\omega^2}{c_{ph}^2} E_1 = -\text{curl}/\text{curl} \frac{\hat{\epsilon} - \bar{\epsilon}}{\bar{\epsilon}} E_0. \quad (9.2)$$

Here E_0 is the field of the particle in a homogeneous medium of dielectric permittivity $\bar{\epsilon}$.¹³³ The solution of Eq. (9.2) at large distances from the specimen has the form

$$E_1 = |k(kG)|, \quad (9.3)$$

where

$$G = \frac{e^{ikR}}{4\pi R} \int \frac{\hat{\epsilon} - \bar{\epsilon}}{\bar{\epsilon}} E_0 e^{-ikr} dr.$$

Here R is the distance from the crystal to the point of observation; the integration is performed over the entire crystal. In the case $v < c_{ph}$, the number of photons emitted per unit solid angle and unit frequency interval is

$$\frac{d^2 N}{d\Omega d\omega} = \frac{e^2}{\hbar c} \frac{\omega \bar{\epsilon} \delta^2}{4\pi^2 v^2} \left[1 - J_0 \left(\frac{\beta \sin \theta_e}{\sqrt{1 - \beta^2}} \right) \right]^2 \frac{1 + \cos^2 \theta_e}{\sin^2 \theta_e} \frac{\sin^2(\alpha L)}{\alpha^2}. \quad (9.4)$$

Here we have $\alpha = (\omega/v)(1 - \beta \cos \theta_e) - \tau$, $\beta = v/c_{ph}$, and e is the charge of the particle. The last factor in (9.4) transforms into a delta-function as $L \rightarrow \infty$ and determines the angular and frequency characteristics.

We can also easily find from (2.2) and (9.3) that the radiation is elliptically polarized. Here one of the axes of the polarization ellipse lies in the k, v plane, while the ratio of axes is $\cos \theta_e$. In particular, the forward and backward radiation is circularly polarized, while at an angle of $\pi/2$ to the trajectory of the particle it is linearly polarized perpendicular to the k, v plane. The intensity of the structural radiation is small in comparison with the Čerenkov radiation ($\sim \delta^2$). However, estimates show that this radiation is ac-

cessible to experimental observation. Actually, for a specimen of thickness $L \sim 10^{-2}$ cm, $p \sim 10^3$ Å, $\delta \sim 10^{-2}$, and $\beta \sim 0.9$, the number of photons emitted by one particle amounts to $N \approx 10^{-3}$.

In the treatment performed above, we have not accounted at all for the effect of the periodicity of the CLC on the propagation of the emitted photons. For certain frequencies and directions of emission, the periodicity proves to be extremely significant. Such frequencies and directions are those for which diffractive scattering of light by the structure of the CLC occurs. The effect of periodicity on Vavilov-Čerenkov radiation in a CLC will be further taken into account below.

b) Vavilov-Cherenkov radiation in cholesteric liquid crystals^{127,129,130}

As we shall show, besides the well known cone of Čerenkov radiation, a second cone of coherent radiation is realized in a CLC that stems from diffraction of light in the CLC. For each direction in the second, diffractive cone, radiation is emitted at its own frequency ω_B , or more exactly, in the frequency range $\Delta\omega/\omega_B \sim \delta$. The radiation in the stated frequency range for the Čerenkov and diffractive cones is not polarized linearly as is usual in Čerenkov radiation. Rather, in the general case it is elliptically polarized, with polarization characteristics depending on both the direction of emission and on the thickness of the crystal. The ratio of intensities of the radiation in these cones depends on the thickness of the crystal and can be of the order of unity for thick enough crystals.

Let us study the Vavilov-Čerenkov radiation of a particle moving uniformly in a direction that makes an arbitrary angle with the optic axis of the CLC. We shall assume that the velocity v of the particle satisfies the condition $v > c_{ph}$. Generally, in addition to the wave lying on the Čerenkov cone and having the wave vector k_0 , in a CLC a wave of the same frequency is generated that is related to it by the Bragg condition. Hence we shall seek the radiation field of the particle (its temporal and spatial Fourier components) as a superposition of two waves $E(k_0) \equiv E_0$ and $E(k_1) \equiv E_1$, where k_0 lies in the region of the Čerenkov cone, and $k_1 = k_0 + \tau$. Analogously to (4.4), we obtain from the Maxwell equations the following system of equations:

$$\left(\hat{\epsilon}_0 - \frac{k_0^2 c^2}{\omega^2} \right) E_0 + \hat{\epsilon}_1 E_1 = \frac{ie\delta(\omega - k_0 v)}{2\pi^2 \omega} \left[v - \frac{k_0(k_0 v)}{k^2} \right], \quad (9.5)$$

$$\hat{\epsilon}_1 E_0 + \left(\hat{\epsilon}_0 - \frac{k_1^2 c^2}{\omega^2} \right) E_1 = 0.$$

The radiation field in a specimen of finite dimensions, which must be determined in order to find the radiation from the crystal, amounts to a superposition of the solutions of the system (9.5) with the solutions of the homogeneous system (4.4), which are obtained from (9.5) by dropping the right-hand sides. The coefficients in this superposition are determined from the boundary conditions.

The frequencies ω_B and the directions k_0 and k_1 in whose vicinity the two waves are emitted are determined by the conditions

$$\omega_B - k_0 v = 0, \quad |k_0 + \tau| = |k_0| = \frac{\omega_B}{c_{ph}}. \quad (9.6)$$

When we neglect the frequency-dependence of $\bar{\epsilon}$, we get from (9.6) the following relationship of ω_B to the direction of the vector k_0 lying on the surface of the Čerenkov cone:

$$\omega_B = \frac{\tau c_{ph}}{2 \cos k_0 \tau}. \quad (9.7)$$

The frequencies ω_B defined by the relationship (9.6) and the corresponding wave vectors k_0 and k_1 can also be found by using a geometric construction (Fig. 17).

The maxima of the amplitudes E_i as functions of k_0 in the solution of the inhomogeneous system (9.5) that correspond to the emission maxima are reached at the points that correspond to a minimum of the modulus of D_1 , which is the determinant of the matrix of system (9.5). Solution of the boundary problem shows that two cases are characteristic of the observed pattern of radiation from the crystal. In one case beating occurs (with the thickness of the crystal or the velocity of the particle being varied) in the intensity of the waves E_0 and E_1 . In the other case we have a strong dependence of E_0 and E_1 on the frequency in the range $|\nu| \lesssim \delta$, where $\nu = (\omega/\omega_B) - 1$. To illustrate the statements that we have made, we analyze below a case that allows simplification of the general expressions.

Let us examine a situation in which a particle is moving at an angle to the surface or a specimen of thickness L , whose optic axis is perpendicular to the surface, while a direction k_0 lies on the surface of the Čerenkov cone and coincides with the direction of the optic axis of the CLC. We find for this direction by using (9.7) that $\omega_B = \tau c_{ph}/2$, while the vector k_1 also lies along the axis of the CLC, but on the side opposite to k_0 . Near the obtained values of ω_B , k_0 , and k_1 , the dynamic system (9.5) breaks down into two independent systems of two equations, one of which describes the diffracted circularly polarized wave n_+ , while the other describes the undiffracted one n_- .¹²⁹ For n_- the matrix is diagonal and diffractive scattering is absent. For n_+ the solution of the dynamical system taking the boundary conditions into account can be written in explicit form.¹²⁹ The amplitudes E_i depend sharply on the frequency and they reach a minimum at $\nu = 0$ and a maximum at $|\nu| \approx \delta/2$. The intensity of the radiation emerging from the crystal is given by the expressions

$$I_0 = I_c \frac{\nu^2 - (1/2)(\delta/2)^2 [1 + (\sin 2\kappa L q / 2\kappa L q)]}{\nu^2 - (\delta/2)^2 \cos^2 \kappa L q},$$

$$I_1 = I_c \frac{(1/2)(\delta/2)^2 [1 - \sin 2\kappa L q / 2\kappa L q]}{\nu^2 - (\delta/2)^2 \cos^2 \kappa L q}. \quad (9.8)$$

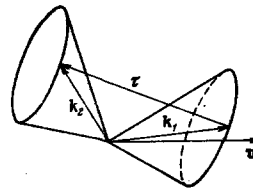


FIG. 17. Diffraction cone of Čerenkov radiation in a CLC. The wave vectors of the radiation in the diffraction cone k_1 are defined by the condition $k_1 = k_0 + \tau$, where k_0 lies in the Čerenkov cone.

Here

$$I_c = \frac{e^2 \omega_B \sin^2 k_0 \tau}{2\pi c^3 \text{ph} \cos k_0 \tau}$$

is the spectral density of Čerenkov radiation in a homogeneous specimen of thickness L , and we have $q = \sqrt{\nu^2 - (\delta/2)^2}$.

Thus, as the presented formulas imply, the radiation intensity in the Čerenkov (forward) and diffracted (backward) cones as a function of the frequency near ω_B undergoes sharp changes in the region $|\nu| \lesssim \delta$ in connection with the diffractive scattering of the polarization n_{\cdot} . When $\nu \gg \delta$ it approaches I_c in the Čerenkov cone and zero in the diffraction cone. As for the polarization of the radiation, in the Čerenkov cone, just like the intensity, it varies with the frequency, approaching linearity when $\nu \gg \delta$. In the diffracted cone the radiation has the polarization n_{\cdot} .

As the treatment performed above implies, the periodicity of the crystal qualitatively alters the nature of the Vavilov-Čerenkov radiation in the neighborhood of particular frequencies and directions. The size of the angular and frequency regions for the cited changes is of the order of δ (a characteristic value of δ is $\sim 10^{-1} - 10^{-2}$). That is, the changes, not only in the angle-integrated but also in the differential characteristics of the radiation, though small, are quite accessible to experimental study.

10. CONCLUSION

This review has concentrated its basic attention on the optical properties of perfect cholesteric crystals. The stated restriction is not fortuitous—in general outline it reflects the state of the theory. As we have noted, on the whole only the optical properties of perfect CLCs are fully understood and amenable to quantitative description. As is evident from what we have presented, theory is ahead of experiment in this field. Here testing a number of theoretical results experimentally and drawing a conclusion concerning their degree of significance for applications yet remain to be done. As for nonideal structures, the situation is the reverse. They are the most accessible object for study, yet the progress of theory in describing the corresponding optical experiments is substantially more modest, and theory lags behind experiment.

In essence, the theoretically studied situations (perfect and mosaic CLCs) constitute two idealized limiting cases, while the actual situation lies between them and can correspond to these limits only to a greater or lesser degree. Yet the existence of theoretical results for only the two discussed limiting models can already be employed to obtain quantitative information from the experimental data, both regarding the structural parameters and the degree of ideality of a CLC. One can get such information on the degree of perfection of a specimen by comparing the observed optical properties of the CLC (e.g., the frequency widths of the regions of selective reflection or the polarization characteristics) with the theoretical results for the models under discussion.

Up to now primarily a particular geometry of scattering (a plane configuration with the optic axis perpendicular to the surface) has been studied theoretically and experimentally. It is also of interest to study the optics of a CLC for other scattering geometries, e.g., specimens with optic axes not perpendicular to the surface. Such situations can be realized through the orienting effect of boundaries or by applying external fields to the CLC.

In line with what we have said, it is pertinent to mention again certain problems of the optics of CLCs that have not yet been studied experimentally, as well as problems whose theoretical development is being stimulated by present experimental potentialities. For perfect CLCs the case of oblique incidence has been insufficiently studied experimentally, and in particular, the higher orders of reflection as well as the polarization characteristics of the scattering. Of considerable interest are optical studies of the dynamics, in particular, of relaxation processes in CLCs, investigations of the optics of absorbing CLCs, and of the Borrmann effect manifested here (suppressed absorption). A theoretical description is lacking of representing individual defects and singular lines (e.g., disinclinations) in CLCs. Radiation from charged particles in CLCs has not yet been studied at all experimentally. The optics and electrooptics of chiral smectic liquid crystals also require further experimental and theoretical study.

We note also the urgency of theoretical development of problems of light scattering in CLCs for some lines of study not touched on here: scattering and rotation of the plane of polarization near phase transition points,^{134-137,124} and nonlinear optics of CLCs.¹³⁸⁻¹⁴⁰

It is useful also to bear in mind that many theoretical results that have been obtained in CLC optics can be used to describe the diffraction of radiation of different types by perfect and imperfect periodic structures (scattering of Mössbauer radiation¹⁴¹ and of neutrons¹⁴² by magnetically ordered structures, and the optics of magnetic helical structures,¹⁴³⁻¹⁴⁵ etc.).

We stress that the above-presented diffractive approach to CLC optics gives an adequate and simple physical interpretation of the discussed phenomena. Hence we can hope that its further development will prove useful for CLC optics, all the more in that the quantitative results of this approach have also proved to be practically exact in typical situations.

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