sager coefficient should in our case be taken into account at temperatures

$$\tau_v \leqslant \left[\frac{AC}{T_c^2} \left(\frac{dT_c}{dC} \right)^2 \right]^{1/v} \approx 10^{-3,2}.$$

Thus, the estimate obtained for the reduced temperatures on the basis of the isomorphism theory of critical phenomena agrees with the characteristic temperatures shown in Fig. 5.

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Contribution to the theory of the optical properties of imperfect cholesteric liquid crystals

V. E. Dmitrienko and V. A. Belyakov

All-Union Research Institute of Physicotechnical and Radio Measurements

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A theory of the optical properties of imperfect (mosaic) cholesteric liquid crystals (CLC) is developed. The optical characteristics are determined on the basis of the transport equations for the light-polarization tensor. The use of these equations is made possible by the statistical character of the imperfections of the mosaic CLC. The case of light propagation along the optical axis is considered in detail. It is shown that the optics of imperfect CLC is subject to qualitative singularities in comparison with the case of ideal CLC. These are, in particular, the polarization of the light, the broadening of the frequency regions of the selective reflection, the independence of the specific rotatory power on the sample thickness, and the selective attenuation of light circularly polarized in either direction. It is noted that the equations obtained in the paper for the polarization tensor can be useful for a description of the scattering of radiation of different types from imperfect crystals.

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INTRODUCTION

Many experimental and theoretical studies have been made of the optics of cholesteric liquid crystals (CLC) (see, e.g.,[1-8]). It has been well established that the unusual character of the optical properties of CLC is due to diffraction of light by their periodic structure. There is, however, a wide gap between the theoretical and experimental papers. Whereas in experiment one deals usually with crystals that are non-ideal to a greater or lesser degree, the theoretical papers consider almost exclusively the optics of an ideal planar cholesteric structure. It appears that the only theoretical relation used for the interpretation of measurements made on non-ideal CLC is the improved Bragg formula,[3,4] which connects the period of the cholesteric helix with the wave length of the diffraction-reflected light and with the angles of incidence and reflection relative to the CLC surface.

Yet there are many problems in the optics of imperfect cholesteric crystals, of importance both from the experimental and the fundamental points of view, which have not been dealt with theoretically. Foremost among them are the dependence of the light reflection and transmission coefficients on the degree of perfection of the cholesteric samples, the influence of the imperfections on the polarization characteristics of the light and on the rotation of the plane of polarization, and others. Naturally, in these cases the phenomena are likewise determined by the diffraction of the light in the CLC, but to answer the foregoing questions it is necessary in most

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cases to go outside the framework of the kinematic approach mentioned above, which leads to the modernized Bragg formula.[3,4]

The purpose of the present paper is to fill in part the indicated gap between theory and experiment and to consider the optical properties of a mosaic non-ideal CLC, i.e., one consisting of a large number of crystallites, small regions with perfect structure, whose orientation and arrangement can differ slightly from the corresponding values in an ideal CLC.

The theoretical treatment of the optical properties of CLC with imperfections of the above type is facilitated by the fact that in CLC with these imperfections there is effectively no coherence in the scattering of the light by the various crystallites. This simplifies the allowance for multiple scattering of light and makes it possible, by generalizing the known Zahariazan equations for x-ray scattering,[7] to obtain results outside the framework of the kinematic approximation. It turns out then that the observed optical properties of imperfect CLC differ substantially from the properties of ideal planar structures. These differences lie not only in the fact that the relations known for ideal structures are not suitable for a quantitative description of imperfect CLC, but also in the fact that a number of qualitatively new effects appear in the optics of imperfect structures. For example, the depolarization of the light in the sample, a different dependence of the angle of rotation of the light-polarization plane on the thickness of the sample, qualitative singularities in circular dichroism, etc.

In Secs. 1–3 we present a brief derivation of the equations that describe the optical properties of mosaic CLC, and Secs. 4 and 5 deal with the use of these equations for the description of the optical properties of a CLC in which light propagates along the optical axis.

1. FUNDAMENTAL EQUATIONS

We consider the scattering and passage of light through a mosaic CLC sample in the form of a plane-parallel plate (see Fig. 1). We assume that the light frequency and the wave incidence angle θ are such that the orientations of the individual crystallites making up the mosaic sample correspond to Bragg scattering of light by them, or are close to the Bragg condition. We assume also that the dimensions of the individual crystallites are small enough to make the attenuation and reflection of the wave by diffraction scattering by an individual crystallite is small. This, of course, does not mean that the attenuation and reflection of the wave by the entire sample will also be small. Moreover, we are interested primarily in a situation wherein the corresponding quantities are not small for the entire sample, meaning under our assumption that the sample consists of a very large number of crystallites.

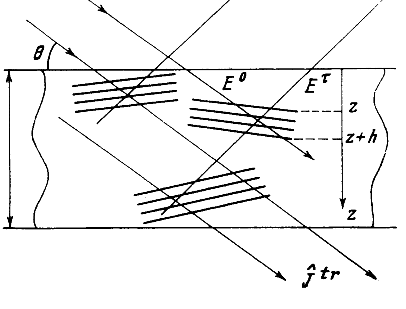

FIG. 1. Schematic representation of mosaic CLC.

Just as in the case of ideal CLC, we use the two-wave approximation, i.e., we represent the wave field in the crystal as a superposition of two waves:

$$\mathbf{E}(\mathbf{r})=\mathbf{E}^0(z)e^{i\mathbf{k}^0\mathbf{r}}+\mathbf{E}^\tau(z)e^{i\mathbf{k}^\tau\mathbf{r}}, \tag{1}$$

where k^0 is the wave vector of the incident wave and $\mathbf{k}^\tau=\mathbf{k}^0+\boldsymbol{\tau}$ is the wave vector of the diffracted wave (here $\boldsymbol{\tau}$ is the CLC reciprocal-lattice vector), and the wave amplitudes $\mathbf{E}^0$ and $\mathbf{E}^\tau$ are functions of the penetration depth z (z is the coordinate normal to the surface). It is important in the following analysis that the field at a certain point of the sample or outside the sample is a superposition of the fields of the primary wave and of the waves that are due to scattering (including multiple scattering) by individual crystallites and have random phases. The absence of correlation between the phases of the scattered waves is due to the random character of the deviations of the orientations and positions of the individual crystallites in the mosaic crystal from an ideal structure. This means that the scattering by the individual crystallites is effectively incoherent, and to solve our problem we can change over from equations for the fields to equations for quantities that are quadratic in the fields of the incident and diffracted waves. This approach was developed for the description of the diffraction of x rays by mosaic crystals by Zahariazen,[7] who obtained a system of two differential equations that connect the changes of the intensities of the direct and diffracted waves at a certain point with the values of the intensities at the same point of the sample.

In the case of passage of light through mosaic CLC, the system of corresponding equations does not reduce to the equations for the intensities, owing to the complex polarization characteristics of light scattering in CLC, but includes all possible combinations that are quadratic in the amplitudes of the fields (1), i.e., components of the polarization tensors $J^p_{ik}=E^p_i E^{p*}_k$, where $p=0, \tau$; $i,k=1,2$:

$$\begin{aligned} d\hat{J}^0/dz&=\hat{A}^0\hat{J}^0-\hat{B}^0\hat{J}^0+\hat{C}^{0\tau}\hat{J}^\tau,\\ -d\hat{J}^\tau/dz&=\hat{A}^\tau\hat{J}^\tau-\hat{B}^\tau\hat{J}^\tau+\hat{C}^{\tau 0}\hat{J}^\tau. \end{aligned} \tag{2}$$

Explicit expressions for the fourth-rank tensors $\hat{A}$, $\hat{B}$, and $\hat{C}$ in (2) can be obtained either within the framework of a phenomenological approach, in which case they contain certain constants, or on the basis of a concrete model of the mosaic crystal. We shall discuss this question later on, and for the time being consider the physical meaning of the system (2).

The meaning of Eqs. (2) is quite lucid. The first equation describes the change of $\hat{J}^0$ with changing coordinate, consisting of the change of $\hat{J}^0$ because of the diffraction scattering and is described by the operator $\hat{B}^0$, and the change due to scattering from the τ direction to the zero direction and described the operator $\hat{C}^{0\tau}$. The operator $\hat{A}^0$ in this equation describes the polarization-tensor change due to absorption and birefringence of the light.

The operators $\hat{B}^0$ and $\hat{C}^{0\tau}$ are connected with Bragg scattering and vanish in its absence. The form of the operator $\hat{A}^0$ known from the theory of radiative transfer does not depend on the presence or absence of Bragg scattering.

The second equation describes analogously the change of the polarization tensor $\hat{J}^\tau$.

It should be noted that physically the quantities J_{ik}^p in the system (2) are not the exact values of quantities quadratic in the amplitudes, but are values of the corresponding quantities averaged over crystal regions that are small but larger than the dimensions of an individual crystallite. We point out that if $\hat{A}$, $\hat{B}$, $\hat{C}$, and $\hat{J}$ are regarded as scalars, then the system (2) reduces to the equations of Zahariazen.[7]

In view of the difficulty of finding the explicit form of the coefficients in (2) (they depend on the structure and on the dimensions of the crystallites, on their shapes, on the distribution of their orientations in the sample, etc.) it is expedient to regard them as phenomenological parameters of the theory. It is then possible, using general consideration, to establish certain relations between the coefficients, so as to decrease the number of the parameters of the theory.

2. COEFFICIENTS OF THE FUNDAMENTAL SYSTEM

Using the form of the dielectric tensor of the CLC, we can determine the tensor form of the operators $\hat{A}$, $\hat{B}$, and $\hat{C}$, i.e., write down, apart from the coefficients, the corresponding tensors in Eq. (2).

The dielectric tensor of the CLC is given by[1,2]

$$\hat{\varepsilon}=\begin{pmatrix} \varepsilon+\bar{\varepsilon}\delta\cos\tau z & \bar{\varepsilon}\delta\sin\tau z & 0 \\ \bar{\varepsilon}\delta\sin\tau z & \varepsilon-\bar{\varepsilon}\delta\cos\tau z & 0 \\ 0 & 0 & \varepsilon_3 \end{pmatrix}=\bar{\varepsilon}+\frac{\delta\bar{\varepsilon}}{2}\left(\hat{\varepsilon}_0+\hat{\varepsilon}_\tau e^{-i\tau z}+\hat{\varepsilon}_{-\tau}e^{i\tau z}\right) \tag{3}$$

where $\bar{\varepsilon}=(\varepsilon_1+\varepsilon_2)/2$, $\delta=(\varepsilon_1-\varepsilon_2)/(\varepsilon_1+\varepsilon_2)$, ε_1, ε_2 and $\varepsilon_3=\varepsilon_2$ are the principal values of the tensor is the CLC reciprocal-lattice vector, and p is the pitch of the cholesteric helix. The Fourier components $\hat{\varepsilon}_0$, $\hat{\varepsilon}_\tau$, and $\hat{\varepsilon}_{-\tau}$ of the tensor $\hat{\varepsilon}$ are given by the expressions

$$\hat{\varepsilon}_0=\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -2 \end{pmatrix}, \quad \hat{\varepsilon}_\tau=\hat{\varepsilon}_{-\tau}^*=\begin{pmatrix} 1 & i & 0 \\ i & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{4}$$

We use for $\hat{A}$ a general expression known from transport theory[8] and valid if the anisotropy of the dielectric tensor $\hat{\varepsilon}$ is small:

$$A_{iklm}^{0,\tau}=i\varkappa[(\varepsilon_0{}^t)_{il}\delta_{km}-\delta_{il}(\varepsilon_0{}^{t*})_{km}]/\sin\theta. \tag{5}$$

Here $\hat{\varepsilon}_0^t$ is the transverse (relative to the wave vector) part of the zeroth Fourier component of the dielectric tensor, and $\varkappa=(\bar{\varepsilon})^{1/2}\omega/c$ is the average wave vector of the light in the CLC.

To find the form of the operator $\hat{C}^{\tau 0}$ we make use of the fact that the change $\Delta\mathbf{E}^\tau$ of the τ-th wave as a result of the Bragg scattering of the wave $\mathbf{E}^0$ can be expressed with good accuracy in terms of the amplitude for scattering by a single crystallite in the kinematic (Born) approximation.[9,10] We then obtain

$$\begin{aligned}\Delta E_i{}^\tau &\sim \int\{(\mathbf{e}_i{}^\tau(\hat{\varepsilon}-1)\mathbf{e}_k{}^{0*})E_k{}^0\exp[i(\mathbf{k}^\tau-\mathbf{k}^0)\mathbf{r}]\}dV \\ &\sim f_1(\mathbf{e}_i{}^\tau\hat{\varepsilon}_\tau\mathbf{e}_k{}^{0*})E_k{}^0=f_1F_{ik}{}^{\tau 0}E_k{}^0,\end{aligned} \tag{6}$$

where the integration is over the volume of the crystallite, $\mathbf{e}_i^{0\tau}$ are the unit vectors of the 0 and τ waves; $\mathbf{E}^0$ and $\mathbf{E}^\tau$ are the amplitudes of the 0 and τ waves. The small parameter that determines the applicability of expression (6) and restricts the characteristic crystallite dimension h is the dimensionless quantity $\delta\varkappa h\ll 1$ (see, e.g.,[9]). The amplitude of the diffracted wave is much smaller here than that of the incident wave.

In the approximation considered, the scattering amplitude is represented in (6) as a product of two factors. The tensor quantity $F_{ik}^{\tau 0}$ is determined by the CLC structure and is the analog of the structure amplitude in the x-ray scattering. The factor f_1 determines the dependence of the amplitude on the dimensions of the crystallite, its shape, orientation, and others. It is essential for our purposes that f_1 is appreciable only if the Bragg condition is satisfied (i.e., f_1 is a function of the frequency).

The change of the polarization tensor of the τ wave as a result of the arrival from the zero direction is obtained in the following manner:

$$\Delta J_{ik}{}^\tau=\Delta E_i{}^\tau\Delta E_k{}^{\tau*}\sim|f_1|^2F_{il}{}^{\tau 0}F_{km}^{\tau 0*}E_l{}^0E_m{}^{0*}. \tag{7}$$

Therefore, by defining f_1 in a way that it contains the factors that are immaterial here, we get

$$C_{iklm}^{\tau 0}=\varkappa\overline{|f_1|^2}F_{il}{}^{\tau 0}F_{km}^{\tau 0*}, \tag{8}$$

where $|\overline{f_1}|^2$ is a quantity obtained from $|f_1|^2$ by averaging over the crystallites. Analogously we have $C_{iklm}^{0\tau}=\varkappa|\overline{f_1}|^2F_{il}^{0\tau}F_{km}^{0\tau*}$. Thus, the tensor properties of the operators $C^{0\tau}$ and $C^{\tau 0}$ are determined entirely by the properties of the structure amplitude F_{il}. The operators $\hat{B}$ are determined similarly in terms of the structure amplitude.

To find $\hat{B}^0$ it is necessary to determine the increment to the amplitude of forward scattering by a single crystallite from the Bragg scattering in the second Born approximation. It can be assumed that it takes the form

$$\Delta E_i{}^0\approx f_2F_{il}{}^{0\tau}F_{lk}{}^{\tau 0}E_k{}^0, \tag{9}$$

where f_2 is a factor analogous to f_1 and reflects the dependence of the increment on the volume, shape, and orientation of the crystallite. Thus, after passing through a single crystallite the wave amplitude can be represented in the form $E_i^0=(\delta_{ik}+f_2F_{in}^{0\tau}F_{nk}^{\tau 0})E_k^0$. Hence

$$\Delta J_{ik}{}^0\sim f_2F_{in}{}^{0\tau}F_{nl}{}^{\tau 0}\delta_{km}E_l{}^0E_m{}^{0*}+f_2{}^*\delta_{il}F_{kn}^{0\tau*}F_{nm}^{\tau 0*}E_l{}^0E_m{}^{0*}, \tag{10}$$

i.e.,

$$B^{0}_{iklm}=\varkappa(\bar{f}_2 F_{in}{}^{0\tau}F_{nl}{}^{\tau 0}\delta_{km}+\bar{f}_2{}^{*}\delta_{il}F_{kn}^{0\tau *}F_{nm}^{\tau 0 *}).$$

For $\hat{B}^{\tau}$ we obtain in similar manner

$$B^{\tau}_{iklm}=\varkappa(\bar{f}_2 F_{in}{}^{\tau 0}F_{nl}{}^{0\tau}\delta_{km}+\bar{f}_2{}^{*}\delta_{il}F_{kn}^{\tau 0 *}F_{nm}^{0\tau *}). \qquad (11)$$

The functions f_1 and f_2 in the operators $\hat{C}$ and $\hat{B}$ in (2) are not independent. It follows from the energy conservation law that $B^{0}_{iilm}=C^{\tau 0}_{iilm}$, whence, using the relation $F^{\tau 0 *}_{km}=F^{0\tau}_{mk}$, we get $2\,\mathrm{Re}\,\bar{f}_2=|\overline{f_1}|^2$. It follows therefore that the function f_2 is expressed in terms of $|\overline{f_1}|^2$, since the real and imaginary parts of f_2 are connected by the dispersion relations.[10]

We have thus determined all the coefficients in Eq. (2) accurate to $|\overline{f_1}|^2$. This quantity, which is a parameter of the theory, can be calculated for simplified models in explicit form.

3. MODEL-DEPENDENT CALCULATION OF THE COEFFICIENTS

To get an idea of the form of f_1 and f_2 and their dependence on the parameters of the mosaic crystal, we obtain their expressions in explicit form for a simple model. We assume that the crystallites are thin plane-parallel plates and differ little in their orientations (see Fig. 1). For a single crystallite we obtain from Maxwell's equations and from (1) and (3) the equations for $\mathbf{E}^0$ and $\mathbf{E}^{\tau}$ (see, e.g.,[11]):

$$\begin{aligned}\frac{d\mathbf{E}^0}{dz}&=-i\frac{\delta\varkappa}{4\sin\theta}\hat{\varepsilon}_0\mathbf{E}^0-i\frac{\delta\varkappa}{4\sin\theta}\hat{\varepsilon}_{-\tau}\mathbf{E}^{\tau},\\ -\frac{d\mathbf{E}^{\tau}}{dz}&=-i\frac{\delta\varkappa}{4\sin\theta}\hat{\varepsilon}_{\tau}\mathbf{E}^0-i\frac{\delta\varkappa}{4\sin\theta}\hat{\varepsilon}_0\mathbf{E}^{\tau}+i\varkappa\alpha\mathbf{E}^{\tau},\end{aligned} \qquad (12)$$

where the parameter $\alpha=(\tau-2\varkappa\sin\theta)/\varkappa$ characterizes the deviation from the Bragg condition (in frequency and in the incidence angle). Assume that only a wave E^0 with amplitude $\mathbf{E}^0(z)$. The amplitudes of the waves $\mathbf{E}^0(z+h)$ and $\mathbf{E}^{\tau}(z)$ that emerge of the crystallite are determined from (12):

$$\begin{aligned}\mathbf{E}^0(z+h)&=\left[1-\delta^2\frac{1+i\alpha\varkappa h-\exp(i\alpha\varkappa h)}{16\alpha^2\sin^2\theta}\hat{\varepsilon}_{-\tau}\hat{\varepsilon}_{\tau}\right]\mathbf{E}^0(z),\\ \mathbf{E}^{\tau}(z)&=\delta\frac{1-\exp(i\alpha\varkappa h)}{4\alpha\sin\theta}\hat{\varepsilon}_{\tau}\mathbf{E}^0(z).\end{aligned} \qquad (13)$$

By determining $\hat{J}^0(z+h)$ and $\hat{J}^{\tau}(z)$ from (13) and recognizing that $\hat{J}^{\tau}(z+h)=0$, we obtain the changes of $\hat{J}^0$ and $\hat{J}^{\tau}$ over the thickness of the crystallite:

$$\begin{aligned}\Delta\hat{J}^0/\Delta z&=-\varkappa\{f_2(\alpha)\hat{\varepsilon}_{-\tau}\hat{\varepsilon}_{\tau}\hat{J}^0+f_2{}^{*}(\alpha)\hat{J}^0\hat{\varepsilon}_{\tau}{}^{+}\hat{\varepsilon}_{-\tau}{}^{+}\},\\ -\Delta\hat{J}^{\tau}/\Delta z&=\varkappa|f_1(\alpha)|^2\hat{\varepsilon}_{\tau}\hat{J}^0\hat{\varepsilon}_{\tau}{}^{+},\end{aligned} \qquad (14)$$

where

$$\begin{aligned}|f_1(\alpha)|^2&=\frac{\delta^2}{8\sin^2\theta}\frac{1-\cos(\alpha\varkappa h)}{\alpha^2\varkappa h},\\ \mathrm{Re}\,f_2&=\frac{1}{2}|f_1|^2,\quad \mathrm{Im}\,f_2=\frac{\delta^2}{16\sin^2\theta}\frac{\alpha\varkappa h-\sin(\alpha\varkappa h)}{\alpha^2\varkappa h}.\end{aligned} \qquad (15)$$

We have thus determined the functions $|f_1|^2$ and f_2 in a simple model. The functions $|f_1|^2$ and $\mathrm{Im}f_2$ have the meaning of the reflecting and rotatory powers of a CLC layer of thickness h. Their dependence on ω is shown dashed in Figs. 2a and 2b, where $\omega_B=c\tau/2\bar{\varepsilon}^{1/2}\sin\theta$. To take into account the presence of the crystallites of varying thickness and orientation, it is necessary to average $|\overline{f_1}|^2$ and f_2 with allowance for the real distributions of the crystallite disorientation angles and of their thicknesses. The averaging smoothes out the functions $|f_1|^2$ and $\mathrm{Im}f_2$, so that $|\overline{f_1}|^2$ and $\mathrm{Im}\bar{f}_2$ take approximately the forms shown by the solid lines in Figs. 2a and 2b.

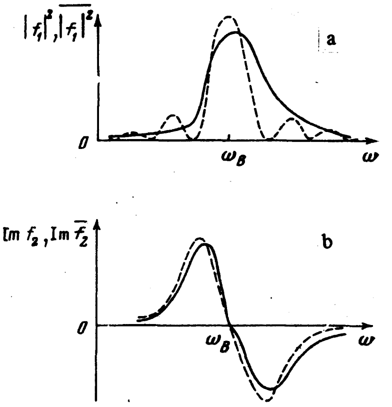

FIG. 2. a) Frequency dependences of the quantities $|f_1|^2$ (dashed) and $|\overline{f_1}|^2$ (solid); b) frequency dependences of $\mathrm{Im}\,f_2$ (dashed) and $\mathrm{Im}\,\bar{f}_2$ (solid).

4. CASE OF NORMAL INCIDENCE

In the case of normal incidence of light on a CLC it is convenient to introduce the circular-polarization unit vectors $\mathbf{e}^{+}$ and $\mathbf{e}^{-}$ (a wave with polarization $\mathbf{e}^{+}$ is diffracted by a perfect CLC, but a wave with polarization $\mathbf{e}^{-}$ is not). In terms of these unit vectors we have

$$\hat{\varepsilon}_{\tau}=\hat{\varepsilon}_{-\tau}=\begin{pmatrix}2&0\\0&0\end{pmatrix}$$

(we do not write here the $\hat{\varepsilon}_{\tau}$ components that will not be needed later on). The system (2) breaks up into several independent systems for the polarization-tensor components. For J^{0}_{11} and J^{τ}_{11} we have

$$\begin{aligned}dJ_{11}{}^{0}/dz&=-(\mu+\mu_d)J_{11}{}^{0}+\mu_d J_{11}{}^{\tau},\\ -dJ_{11}{}^{\tau}/dz&=-(\mu+\mu_d)J_{11}{}^{\tau}+\mu_d J_{11}{}^{0},\end{aligned} \qquad (16)$$

where $\mu_d=4\varkappa|\overline{f_1}|^2$, and the subscript 1 pertains to the polarization unit vector $\mathbf{e}^{+}$ (the subscript 2 pertains to $\mathbf{e}^{-}$), i.e., the system (16) describes waves with $\mathbf{e}^{+}$ polarization that undergo diffractive scattering. In the system (16) and the following we take into account the nondiffractive attenuation of the light in the mosaic CLC (e.g., via scattering by the crystallite boundaries) by introducing the linear absorption coefficient μ.

The propagation of waves with circular polarization $\mathbf{e}^{-}$ is described by the following equations:

$$dJ_{22}{}^{0}/dz=-\mu J_{22}{}^{0},\quad dJ_{22}{}^{\tau}/dz=\mu J_{22}{}^{\tau}. \qquad (17)$$

The off-diagonal elements of the polarization tensors satisfy also the non-coupled equations

$$\begin{aligned}\frac{dJ_{12}{}^{0}}{dz}&=-(\mu+4\varkappa\bar{f}_2)J_{12}{}^{0}, &\frac{dJ_{12}{}^{\tau}}{dz}&=(\mu+4\varkappa\bar{f}_2)J_{12}{}^{\tau},\\ \frac{dJ_{21}{}^{0}}{dz}&=-(\mu+4\varkappa\bar{f}_2{}^{*})J_{21}{}^{0}, &\frac{dJ_{21}{}^{\tau}}{dz}&=(\mu+4\varkappa\bar{f}_2{}^{*})J_{21}{}^{\tau}.\end{aligned} \tag{18}$$

To find the polarization tensors of the transmitted wave and the wave reflected from the crystal, we must add boundary conditions to Eqs. (16)–(18). Namely, on the entrance surface of the crystal (at $z=0$) we put $\hat{J}^0(0)=\hat{J}^{\text{ext}}$, where $\hat{J}^{\text{ext}}$ is the polarization tensor of the wave incident on the crystal, and at the exit surface (at $z=l$) we put, since there is no diffracted wave, $\hat{J}^{\tau}(l)=0$.

We shall not write out the calculations, but present directly the solutions of (16)–(18) with allowance for the boundary conditions.

For the reflected wave we have

$$\begin{gathered}J_{11}^{ref}=J_{11}^{ext}\frac{\mu_d \operatorname{sh}\lambda_1 l}{(\mu+\mu_d)\operatorname{sh}\lambda_1 l+\lambda_1\operatorname{ch}\lambda_1 l},\\ J_{22}^{ref}=J_{12}^{ref}=J_{21}^{ref}=0,\quad \lambda_1=(\mu^2+2\mu\mu_d)^{1/2}.\end{gathered} \tag{19}$$

We obtained a natural result: the only reflected component of the entire incident beam is the one with polarization $\mathbf{e}^{+}$, and the reflected light preserves this polarization. The reflection coefficient for this polarization is simply the coefficient of J_{11}^{ext} in (19). If the absorption is small (i.e., $\lambda_1 l \ll 1$), then

$$J_{11}^{ref}=\frac{\mu_d l J_{11}^{ext}}{(1+\mu_d l)}. \tag{20}$$

We see therefore that the reflection coefficient is appreciable if $\mu_d l>1$. The plot of the reflection from a thick imperfect crystal can be greatly broadened in comparison with the frequency-dependent width of the function $\mu_d=4\varkappa|\bar{f}_1|^2$, which is determined by the dimensions of the crystallites and their angular disorientation.

For the transmitted wave we have

$$J_{11}{}^{tr}=\frac{\lambda_1}{(\mu+\mu_d)\operatorname{sh}\lambda_1 l+\lambda_1\operatorname{ch}\lambda_1 l}J_{11}^{ext}, \tag{21a}$$

$$J_{22}{}^{tr}=J_{22}^{ext}e^{-\mu l}, \tag{21b}$$

$$J_{12}{}^{tr}=J_{21}^{tr\,*}=J_{12}^{ext}\exp[-(\mu+4\varkappa\bar{f}_2)l]. \tag{21c}$$

It is seen from (21a) and (21b) that, owing to diffractive reflection, the intensity of the transmitted wave with polarization $\mathbf{e}^{+}$ decreases more rapidly with increasing thickness than the intensity of the wave with $\mathbf{e}^{-}$ polarization, which undergoes only the usual damping.

Another qualitative distinguishing feature is that the transmitted light is, generally speaking, depolarized. As can be seen from (21), the degree of depolarization of the transmitted light is minimal for a perfectly defined polarization of the incident light (this polarization depends on the thickness of the sample and is determined by the condition $J_{11}^{\text{tr}}=J_{22}^{\text{tr}}$). In this case the emerging light is partially linearly polarized and its degree of polarization is

$$P_{min}=\left(\operatorname{ch}\lambda_1 l+\frac{\mu+\mu_d}{\lambda_1}\operatorname{sh}\lambda_1 l\right)^{1/2}\exp\left[\frac{-(\mu+\mu_d)l}{2}\right]. \tag{22}$$

On the other hand, light with circular polarization $\mathbf{e}^{+}$ or $\mathbf{e}^{-}$ does not become depolarized.

Let us consider by way of example the case of linearly polarized incident light for which the polarization tensor is given by

$$\hat{J}^{ext}=\frac{I^{ext}}{2}\begin{pmatrix}1 & -i\\ i & 1\end{pmatrix}, \tag{23}$$

where I^{ext} is the intensity of the incident light (we recall that we use the circular-polarization unit vectors as the basis).

As already noted, the reflected light is circularly polarized, while the light transmitted through the sample is in the general case partially (elliptically) polarized. The ellipse rotation angle φ at the exit is given by

$$\operatorname{tg}2\varphi=\frac{\operatorname{Re}J_{12}{}^{tr}}{\operatorname{Im}J_{12}{}^{tr}}=\frac{\sin(4\varkappa\operatorname{Im}\bar{f}_2 l)}{\cos(4\varkappa\operatorname{Im}\bar{f}_2 l)}, \tag{24}$$

i.e., $\varphi=2\varkappa l\operatorname{Im}\bar{f}_2$. It is seen that φ is the sum of the angles of rotation by the individual crystallites and is proportional, in contrast to ideal CLC,[2,12] to the crystal thickness. Thus, $2\varkappa\operatorname{Im}\bar{f}_2$ is the rotatory power of an imperfect CLC.

If the absorption coefficient μ is small (so that $\lambda_1 l \ll 1$), then simple expressions are obtained also for the remaining polarization characteristics. For the degree of polarization P we have

$$P=(\mu_d{}^2l^2+4(1+\mu_d l)^2\exp(-\mu_d l))^{1/2}/(2+\mu_d l), \tag{25}$$

for the ratio p of the polarization-ellipse axes we have

$$p=\mu_d l\{2(1+\mu_d l)\exp(-\mu_d l/2)+[\mu_d{}^2l^2+4(1+\mu_d l)^2\exp(-\mu_d l)]^{1/2}\}^{-1}; \tag{26}$$

for the intensity I^{tr} of the transmitted light we have

$$I^{tr}=\frac{I^{ext}}{2}\left(\frac{2+\mu_d l}{1+\mu_d l}\right). \tag{27}$$

The thickness dependence of these quantities is clear (see Fig. 3): At small thicknesses we have $P=1$ and $p=0$, since the incident light is linearly polarized and the influence of the crystal is weak. With increasing l, the degrees of the polarizations first decrease (because of the multiple incoherent scattering) and the transmitted

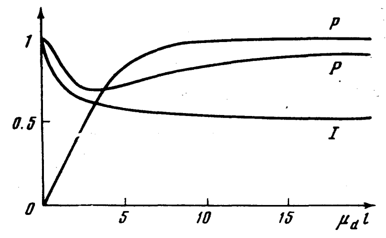

FIG. 3. Dependence of the transmission coefficient $I=I^{\text{tr}}/I^{\text{ext}}$, of the degree of polarization P, and of the ratio of the polarization-ellipse axes p on the sample thickness for linearly incident light.

beam becomes partially polarized, but with further increase of the thickness the degree of polarization again tends to unity, since only e^- polarization passes through a very thick crystal. We recall that polarized light passing through an ideal crystal experiences no depolarization whatever.

5. ATTENUATION OF LIGHT WITH NONDIFFRACTIVE POLARIZATION

It is seen from the foregoing analysis of the case of normal incidence that the e^+ and e^- circular polarizations propagate in the CLC independently, and only the e^+ polarization is reflected. In imperfect CLC, however, there is present a nondiffractive mechanism for the conversion of e^+ polarization into e^- (and vice versa), and leads also to a selective attenuation of the light with the e^- polarization. This conversion is produced, for example, by optical inhomogeneities of the mosaic crystal. The operator $\hat{A}$ (5) introduced above does not take these processes into account and must be modified.

In the case of normal incidence this means that the nonvanishing components of the operator A are, besides A_{1111}, A_{2222}, A_{1212}, and A_{2121}, also A_{1122} and A_{2211}. Solving Eq. (2) with $\hat{A}$ in this form, we obtain in the case of incident e^- polarization

$$J_{22}^{tr}=J_{22}^{ext}\exp(-\lambda_2 l),$$

$$\lambda_2=\{(\mu+\xi)^2+(\mu+\xi)\mu_d+\xi^2-(\mu+\xi)^{1/2}[(\mu+\xi)(\mu_d^2+4\xi^2)+4\mu_d\xi^2]^{1/2}\}^{1/2} \quad (28)$$

(we assume for simplicity that $A_{1122}=A_{2211}=\xi$ and that these terms are small in comparison with all others).

In the selective-reflection region we have $\xi \ll \mu_d$ and $\lambda_2=\mu+\xi$; outside this region $\mu_d \approx 0$ and $\lambda_2=\mu$. Thus, as follows from (28), the attenuation of the wave with e^- polarization should increase as a function of the frequency in the region of the diffraction reflection. Another qualitative manifestation of the optical inhomogeneity of the CLC is the reflected-beam depolarization, but its value is small and is of the order of the smaller of the ratios ξ/μ or ξ/μ_d.

The parameter ξ in (28) can be found from measurements of the optical characteristics of the CLC outside the selective reflection region. We can use for this purpose the fact that the components A_{1122} and A_{2211} describe the depolarization of the direct beam outside the Bragg region. For example, the degree of polarization of light passing through the sample (if the incident light is circularly polarized) is given by the expression

$$P=e^{-2\xi l}. \quad (29)$$

We note that the results of this section agree with[13,14] where selective attenuation of the light with nondiffractive polarization has been observed.

CONCLUSION

We have considered above the optics of CLC using an ideally mosaic crystal as the model. Another case investigated in detail is that of a perfect cholesteric structure.[5,6,12–16] Let us analyze the difference between the results given by the two models and the degree of their agreement with the realized experimental situations. The most substantial differences in the theoretical deductions are observed in the frequency widths of the selective-reflection regions, and also in the polarization characteristics.

In mosaic CLC, the broadening of the frequency domain of the selective reflection increases with thickness. The selective region of a thick crystal is larger than that of a thin crystal having the same degree of perfection, owing to the multiple-scattering processes (20) and (21). In ideal CLC the tendency is reversed.

In a mosaic CLC the specific rotation of the plane of polarization is smaller than in an ideal one, and in contrast to the latter it does not depend on the sample thickness (see (24)). The frequency range of the dispersion of the rotatory power is determined in this case by the crystallite dimensions and, unlike the reflection region it does not change with the crystal thickness.

A qualitative new phenomenon not observed in perfect CLC is the depolarization of light in a mosaic crystal. It must be noted that in a mosaic CLC, besides the usual beam depolarization due to the inhomogeneities, a depolarization is caused by diffractive scattering [Eqs. (22) and (25)].

It is known[1,2] that in the case of propagation along the optical axis of an ideal CLC, light of only one circular polarization is selectively reflected and attenuated. In mosaic CLC, the light with the opposite circular polarization is also subject to selective attenuation (28).

As to the question of the model best suitable for the description of the experimental data, the answer depends on the actual organization of the experiment. In general, the experimental results correspond to situations intermediate between the discussed limiting cases. A comparison of the experimental data with the results of the discussed models can therefore be used to obtain quantitative estimates of the degree of perfection of the CLC, for example from the frequency width of the region of the selective reflection, or from the depolarization of the light.

A comparison of the present results with measurement data shows that the singularities noted by us in the optics of mosaic CLC agree with the experimental results. Thus, it is seen from[13,14] that the experimentally observed frequency width of the selective reflection is larger than the dispersion region of the rotatory power; the measured rotatory power turns out to be smaller than that of an ideal CLC. In the same papers, a frequency-dependent (in the selective-reflection region) attenuation of light with either polarization was observed.

We point out in conclusion that the reported method of describing the diffraction of light in mosaic CLC can be directly used in the theory of diffraction of other types of radiation, such as that of neutrons or of Mössbauer radiation in mosaic magnetically ordered crystals.

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Photogalvanic effect in a crystal with polar axis

V. I. Belinicher, V. K. Malinovskiĭ, and B. I. Sturman

Institute of Automation and Electrometry, Siberian Division, USSR Academy of Sciences

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Photoelectric emf mechanisms governed only by the symmetry of the crystal—by the presence of a polar axis—are proposed. They are based on the asymmetry of the electronic processes—their non-invariance to spatial reflections.

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INTRODUCTION

The photoelectric-emf mechanisms connected with the inhomogeneity of crystals and with the non-uniformity of their illumination are well known.[1] There are, however, experimental data indicating that emf is produced in homogeneous ferroelectrics that are uniformly illuminated.[2,3] The existence of photo-induced currents is due in this case to the crystal symmetry—the presence of a polar axis in the crystal[2]; this effect does not exist in the para-phase. The currents observed in short-circuited samples are small ($j_{\mathrm{ph}} \approx 10^{-8}$–$10^{-9}$ A/cm^2 at $J \sim 1$ W/cm^2), but when isolated crystals are illuminated charge transfer gives rise to blocking fields $E_0 \sim 10^3$–10^5 V/cm.

Consider the expansion of the constant electric current in power of the field of the light wave $\tilde{\mathbf{E}}(\omega)$ and the constant field $\mathbf{E}$:

$$j_i = \sigma_{ik} E_k + \beta_{ikl} E_k E_l + \gamma_{iklm} E_k E_l E_m + \ldots.$$

Here σ_{ik} is the intrinsic conductivity of the crystal, γ_{iklm} is the photoconductivity, and β_{ikl} describes the effect considered by us, namely a current in the absence of a constant field. If the crystal has a polar axis, then we can construct the tensor β_{ikl}:

$$\beta_{ikl} = \alpha c_i \delta_{kl} + \beta c_i c_k c_l + \gamma (c_l \delta_{ik} + c_k \delta_{il}), \tag{1}$$

and the unit vector e specifies the polar direction of the crystal. The quantities $\alpha(\omega)$, $\beta(\omega)$, and $\gamma(\omega)$ should be determined by the microscopic approach. Expression (1) for β_{ikl} is not the most general one, since it does not take into account the singularities of the crystal symmetry. Photocurrent is possible if the crystal symmetry allows the existence of the tensor β_{ikl}.

We propose an elementary theory of the effect. This theory is based on the asymmetry of the elementary electronic processes—their noninvariance to spatial reflections. It is assumed that the dielectric has in its forbidden band an impurity level to depth Δ, from which the photoexcited electrons stem. The electrons in the conduction band will be described by the kinetic equation for the distribution function f_k:

$$\frac{\partial f_{\mathbf{k}}}{\partial t} = I^{i}_{\mathbf{k}} - I^{r}_{\mathbf{k}} + I^{\mathrm{imp}}_{\mathbf{k}} + I^{\mathrm{ph}}_{\mathbf{k}}, \tag{2}$$

where $I^{\mathrm{ex}}_{m}(f_{\mathbf{k}})$ and $I^{r}_{\mathbf{k}}(f_{\mathbf{k}})$ are respectively the electron excitation and recombination rates, while $I^{\mathrm{imp}}_{\mathbf{k}}(f_{\mathbf{k}})$ and $I^{\mathrm{ph}}_{\mathbf{k}}(f_{\mathbf{k}})$ are the integrals of the collisions with the impurities and the phonons.

The idea of the effect is the following: Assume that the

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