Optics of a chiral liquid crystal far from a Bragg resonance

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The problem of propagation of light across a dielectric medium with a helical structure, for example, through a cholesteric or a chiral smectic liquid crystal is considered. The problem is solved for an aribitrary angle between the wave vector and the helix axis far from a Bragg resonance. The solution is obtained to within terms $\sim\varepsilon_a^2$ inclusive, where ε_a is the permittivity anisotropy. A full allowance is made for the influence of the longitudinal components of the electric field. A solution is also given of the electrostatic problem of the macroscopic permittivity of a medium with anisotropically correlated tensor fluctuations.

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1. INTRODUCTION

The optics of chiral liquid crystals exhibiting a helical structure (i.e., cholesteric and some smectic liquid crystals) has been investigated intensively for a fairly long time. We recall here a number of studies in which solutions are given for some important special cases[1–4] (the reader is also referred to general monographs on liquid crystals[5,6] and to a review[7] devoted entirely to the optics of cholesteric liquid crystals). In view of the potential applications, most of the investigations of the optics of cholesteric liquid crystals have been devoted to the processes of a resonant Bragg reflection. Another aspect, the gyrotropy of cholesteric liquid crystals, has been investigated mainly for the propagation of light along the helix axis, when there is no disturbing influence of the birefringence.

We shall consider the optics of a chiral liquid crystal far from a Bragg resonance.

We shall assume that the helical pitch is not very large, so as to avoid the regime in which changes in the polarization follow adiabatically those in the orientation of the director (Mauguin limit). We shall use the method of expansion in powers of a small parameter $\varepsilon_a/\bar{\varepsilon}$, representing the relative anisotropy of the permittivity tensor. In the case of a cholesteric this problem, was analyzed fully by Kats with the same accuracy using perturbation theory.[3] Unfortunately, it is extremely difficult to use the results of Kats because of a large number of misprints. Moreover, the equations for the field components E_x and E_y, where z is directed along the helix axis, are used in Ref. 3. This method is suitable only for a cholesteric with a specific type of the permittivity tensor ($\varepsilon_{xz}=\varepsilon_{yz}=0$). We shall solve the Maxwell equations in a covariant vector form, which makes it possible to consider the problem in principle also for smectics.

2. MAXWELL EQUATIONS AND FORM OF TENSOR $\hat{\varepsilon}$

The Maxwell equations for a monochromatic $\exp(-i\omega t)$ field are of the form

$$ip\mathbf{D}(\mathbf{r})=\operatorname{rot}\mathbf{H},\quad ip\mathbf{H}=\operatorname{rot}\mathbf{E},$$
$$D_i=\varepsilon_{ik}(\mathbf{r})E_k,\quad p=\omega/c. \tag{1}$$

Since we aim to allow accurately for the transverse and longitudinal components of the fields, we shall find it more convenient to use the induction vector $\mathbf{D}(\mathbf{r})$, for which we obtain from the system (1) the exact equality $\operatorname{div}\mathbf{D}(\mathbf{r})=0$, and not that containing the vector $\mathbf{E}(\mathbf{r})$. The permittivity tensor will be considered in the form (see Ref. 7)

$$\hat{\varepsilon}=\sum_{s=-2}^{2}\hat{\varepsilon}_s e^{is\mathbf{qr}},\quad \mathbf{qr}\equiv qz. \tag{2}$$

Here, $\mathbf{q}=q\mathbf{e}_z$ is the "wave vector" of the helix; the medium on the right differs from that on the left by the sign of the parameter q. The tensors $\hat{\varepsilon}_{\pm1}$, $\hat{\varepsilon}_{\pm2}$, and $\hat{\varepsilon}_0$ are given by

$$\begin{aligned}(\varepsilon_0)_{xx}&=(\varepsilon_0)_{yy}=\tfrac{1}{2}(\varepsilon_1+\varepsilon_2\cos^2\theta+\varepsilon_3\sin^2\theta),\\ (\varepsilon_0)_{zz}&=\varepsilon_2\sin^2\theta+\varepsilon_3\cos^2\theta,\\ (\varepsilon_0)_{yx}&=(\varepsilon_0)_{yz}=(\varepsilon_0)_{zx}=0,\\ \hat{\varepsilon}_1&=\hat{\varepsilon}_{-1}{}^{*}=\tfrac{1}{4}(\varepsilon_2-\varepsilon_3)\hat{\sigma}_1\sin 2\theta,\\ \hat{\varepsilon}_2&=\hat{\varepsilon}_{-2}{}^{*}=\tfrac{1}{4}(\varepsilon_1-\varepsilon_2\cos^2\theta-\varepsilon_3\sin^2\theta)\hat{\sigma}_2,\end{aligned} \tag{3}$$

$$\hat{\sigma}_1=\begin{pmatrix}0&0&i\\0&0&1\\i&1&0\end{pmatrix},\quad \hat{\sigma}_2=\begin{pmatrix}1&-i&0\\-i&-1&0\\0&0&0\end{pmatrix}.$$

Here, ε_1, ε_2, and ε_3 are the principal values of the local tensor $\hat{\varepsilon}(\mathbf{r})$ and they are independent of $\mathbf{r}$; θ is the angle between the direction of z and the principal (third) axis of the local tensor, which is also independent of $\mathbf{r}$. The second axis of the local tensor is always in the (x,y) plane. The special case of a cholesteric can be obtained from Eqs. (2) and (3) by substituting $\theta=0$ and $\varepsilon_2=\varepsilon_3$; we then have $\hat{\varepsilon}_1=\hat{\varepsilon}_{-1}=0$. The spatial average (i.e., averaged over several periods of the helix) value of the tensor $\hat{\varepsilon}(\mathbf{r})$ is $\hat{\varepsilon}_0$. Therefore, if it were possible to use the space-averaged tensor $\hat{\varepsilon}$, the optical properties would have corresponded to those of a uniaxial crystal (with the optic axis along the helix axis) having the following refractive indices for the ordinary (o) and extraordinary (e) waves:

$$\begin{aligned}n_o&=[\tfrac{1}{2}(\varepsilon_1+\varepsilon_2\cos^2\theta+\varepsilon_3\sin^2\theta)]^{1/2},\\ n_e&=[\varepsilon_2\sin^2\theta+\varepsilon_3\cos^2\theta]^{1/2}.\end{aligned} \tag{4}$$

In this primitive approach the dependence of the length of the wave vector k^2 on the angle φ, where $(\mathbf{ke}_z)=k\cos\varphi$, would be

$$k_o{}^2=p^2n_o,\quad k_e{}^2=p^2[n_o{}^{-2}\cos^2\varphi+n_e{}^{-2}\sin^2\varphi]^{-1}. \tag{5}$$

On the other hand, if the problem is considered in terms

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of the induction $\mathbf{D}$, it is convenient to use a tensor $\hat{\eta}$ which is the reciprocal of $\hat{\varepsilon}$:

$$\hat{\eta}(\mathbf{r})=\sum_{s=-2}^{2}\hat{\eta}_s e^{isqr},$$

$$(\hat{\eta}_0)_{xx}=(\hat{\eta}_0)_{yy}=\frac{1}{2}\left[\frac{1}{\varepsilon_2\varepsilon_3}(\varepsilon_2\sin^2\theta+\varepsilon_3\cos^2\theta)+\frac{1}{\varepsilon_1}\right], \tag{6}$$

$$(\hat{\eta}_0)_{ik}=0,\quad i\neq k;\quad (\hat{\eta}_0)_{zz}=\frac{1}{\varepsilon_2\varepsilon_3}(\varepsilon_2\cos^2\theta+\varepsilon_3\sin^2\theta);$$

$$\hat{\eta}_1=\hat{\eta}_{-1}^{*}=-\frac{1}{4\varepsilon_2\varepsilon_3}(\varepsilon_2-\varepsilon_3)\hat{\sigma}_1\sin 2\theta;$$

$$\hat{\eta}_2=\hat{\eta}_{-2}^{*}=-\frac{1}{4}\left[\frac{1}{\varepsilon_2\varepsilon_3}(\varepsilon_2\sin^2\theta+\varepsilon_3\cos^2\theta)\frac{1}{\varepsilon_1}\right]\hat{\sigma}_2.$$

If it were possible to use the spatial-average value of the tensor $\hat{\eta}(\mathbf{r})$, the optical properties of the medium would have corresponded to a uniaxial crystal width

$$n_o=[(\eta_0)_{xx}]^{-1/2},\quad n_e=[(\eta_0)_{zz}]^{-1/2}. \tag{7}$$

We shall introduce

$$\varepsilon_1=\varepsilon_2(1+\mu_1),\quad \varepsilon_3=\varepsilon_2(1+\mu_3).$$

We can now readily show that Eqs. (7) and (4) give results which are the same to within terms of the first order with respect to μ_1 and μ_3, but differ in respect of the terms $\sim\mu_1^2$, $\mu_1\mu_3$, and μ_3^2. We shall find later the explicit analytic expressions for the two eigenvalues k^2 as a function of φ accurate to within terms up to the order of μ^2 inclusive. With precision to the first order in μ, these expressions are identical with those obtained from Eqs. (4) and (5) or from Eqs. (7) and (5). On the other hand, in the case of terms of the order of μ^2 it is found that not only each of the expressions in Eqs. (4) and (7) gives an incorrect result, but the functional dependence of the (5) type does not hold. This follows readily if only from the fact that according to Eq. (5) the angle $\varphi=0$ (propagation along the axis) corresponds to $k_o^2=k_e^2$. On the other hand, it is well known that the gyrotropy appears precisely in the case where light propagates along the axis of a cholesteric liquid crystal, i.e., in this case we can expect splitting of circularly polarized waves:

$$k_L^2-k_R^2\sim\varepsilon_a^2(\omega/c)^5[q(k^2-q^2)]^{-1}.$$

Less evident is the conclusion derived below on the presence of corrections of the order of $\sim\varepsilon_a^2$ also in the case of large angles φ.

3. SEPARATION INTO TRAVELING AND VIRTUAL WAVES

The complete induction vector $\mathbf{D}(\mathbf{r})$ of Eq. (1) is described by

$$\operatorname{rot}\operatorname{rot}(\hat{\eta}(\mathbf{r})\mathbf{D}(\mathbf{r}))-p^2\mathbf{D}(\mathbf{r})=0. \tag{8}$$

The vector $\mathbf{D}(\mathbf{r})$ will be sought in the form of the main term representing a traveling wave $D\exp(i\mathbf{kr})$ and small corrections representing virtual waves $\mathbf{B}_{\pm1}$ and $\mathbf{B}_{\pm2}$, which are of the first order in $\eta_{\pm1}$ and $\eta_{\pm2}$, respectively:

$$\mathbf{D}(\mathbf{r})=e^{i\mathbf{kr}}\left(\mathbf{D}+\sum_{s=-2}^{2}\mathbf{B}_s e^{isqr}\right). \tag{9}$$

The vectors $\mathbf{D}$ and $\mathbf{B}_s$ on the right-hand side of Eq. (9) are understood to be independent of the coordinates, and $\mathbf{B}_0\equiv0$. We next substitute Eqs. (9) and (6) into Eq. (8) and equate to zero separately each term of the $\exp(i\mathbf{kr}+is\mathbf{qr})$ type with $s=-2,\ -1,\ 0,\ 1$, and 2, ignoring all the higher spatial harmonics. Moreover, we neglect the excitation of various spatial harmonics of the second order in η_s, because the reaction of the main wave would appear only in the μ^3 order. Therefore, in the $s\neq0$ case, we have

$$\operatorname{rot}\operatorname{rot}[\hat{\eta}_0\mathbf{B}_s e^{i(\mathbf{k}+s\mathbf{q})\mathbf{r}}]-p^2\mathbf{B}_s e^{i(\mathbf{k}+s\mathbf{q})\mathbf{r}}=-\operatorname{rot}\operatorname{rot}[\hat{\eta}_s\mathbf{D}e^{i(\mathbf{k}+s\mathbf{q})\mathbf{r}}]. \tag{10}$$

Since we are considering $\mathbf{B}_s$ as corrections to $\mathbf{D}$ linear in μ, the tensor $\hat{\eta}_0$ on the left-hand side of Eq. (10) can be replaced with satifactory precision by $\varepsilon^{-1}\delta_{ik}$, where ε is any of the values ε_1, ε_2, and ε_3. In all those cases when with satisfactory accuracy we can take any of the values ε_1, ε_2, and ε_3, we shall write ε without a circumflex (i.e., we shall assume it to be a pure number) and without subscript. It then follows from Eq. (10) that

$$\mathbf{B}_s=[\varepsilon^{-1}(\mathbf{k}+s\mathbf{q})^2-p^2]^{-1}[(\mathbf{k}+s\mathbf{q})[(\mathbf{k}+s\mathbf{q})(\hat{\eta}_s\mathbf{D})]]. \tag{11}$$

The corrections to the induction $\mathbf{B}_s$ are found to be purely transverse, as expected, because of the exact equality $\operatorname{div}\mathbf{D}(\mathbf{r})=0$. However, in this case electric field vector $\mathbf{E}(\mathbf{r})$ has also longitudinal components which we shall allow for automatically. Without loss of generality, we can assume that

$$\mathbf{k}=k(\mathbf{e}_z\cos\varphi+\mathbf{e}_x\sin\varphi),\quad k=p\varepsilon^{1/2}+O(\mu). \tag{12}$$

Then, with satisfactory accuracy the denominators in Eq. (11) can be written in the form

$$\varepsilon^{-1}(\mathbf{k}+s\mathbf{q})^2-p^2=s\varepsilon^{-1}q(sq+2p\varepsilon^{1/2}\cos\varphi). \tag{13}$$

4. FRESNEL EQUATION FOR THE WAVE VECTOR AND FOR POLARIZATION EIGENSTATES

The equation for $\mathbf{D}\exp(i\mathbf{kr})$ is of the form

$$\operatorname{rot}\operatorname{rot}[\hat{\eta}_0\mathbf{D}e^{i\mathbf{kr}}]-p^2\mathbf{D}e^{i\mathbf{kr}}=-\operatorname{rot}\operatorname{rot}\left[e^{i\mathbf{kr}}\sum_{s\neq0}\hat{\eta}_{-s}\mathbf{B}_s\right]. \tag{14}$$

Substituting Eq. (11) in Eq. (14), we obtain a linear homogeneous equation for $\mathbf{D}$:

$$\{(k^2\delta_{ij}-k_ik_j)[(\eta_0)_{ik}+\psi_{jk}]-p^2\delta_{ik}\}D_k=0. \tag{15}$$

In the above equation we have introduced

$$\psi_{jk}=\sum_{s\neq0}[s\varepsilon^{-1}q(sq+2p\varepsilon^{1/2}\cos\varphi)]^{-1}$$

$$\times\{(\eta_{-s}(\mathbf{k}+s\mathbf{q}))_j(\bar{\eta}_s(\mathbf{k}+s\mathbf{q}))_k-(\mathbf{k}+s\mathbf{q})^2(\eta_{-s}\eta_s)_{jk}\}. \tag{16}$$

The vector $\mathbf{D}$ satisfies the condition of transverseness $(\mathbf{D}\cdot\mathbf{k})=0$ and, therefore, it can be written in the form

$$\mathbf{D}=\mathbf{e}_yD_y+\mathbf{e}_vD_v;\quad \mathbf{e}_v=\mathbf{e}_x\cos\varphi-\mathbf{e}_z\sin\varphi. \tag{17}$$

Then, the system (15) transforms into

$$\begin{cases}[k^2(\mathbf{e}_y\hat{\eta}_0\mathbf{e}_y)+p^2\varepsilon(\mathbf{e}_y\hat{\psi}\mathbf{e}_y)-p^2]D_y+p^2\varepsilon(\mathbf{e}_y\hat{\psi}\mathbf{e}_v)D_v=0,\\ p^2\varepsilon(\mathbf{e}_v\hat{\psi}\mathbf{e}_y)D_y+[k^2(\mathbf{e}_v\hat{\eta}_0\mathbf{e}_v)+p^2\varepsilon(\mathbf{e}_v\hat{\psi}\mathbf{e}_v)-p^2]D_v=0.\end{cases} \tag{18}$$

In the derivation of the system (18) we have allowed for the fact that $(\mathbf{e}_y\,\hat{\eta}_0\mathbf{e}_v)=(\mathbf{e}_v\,\hat{\eta}_0\mathbf{e}_y)=0$; moreover, in terms $\sim\psi$ the factor k^2 can be replaced with satisfactory accuracy by

$p^2\varepsilon$. Equating the determinant of the system (18) to zero, we obtain the following equation for the eigenvalues of k^2:

$$[k^2-k_o^2(\psi)]\,[k^2-k_e^2(\varphi)]-{}^1/_4A^2(\varphi)[k_o^2(\varphi)-k_e^2(\varphi)]^2=0, \quad (19)$$

$$k_o^2(\varphi)=p^2/(\eta_0)_{yy}-p^2\varepsilon^2\psi_{yy}; \quad k_e^2(\varphi)=p^2/(\eta_0)_{vv}-p^2\varepsilon^2\psi_{vv}, \quad (20)$$

$$A^2(\psi)=4|\psi_{yv}|^2[(\eta_0)_{vv}-(\eta_0)_{yy}]^{-2}. \quad (21)$$

The solution of Eq. (19) is

$$(k^2)_{1,2}={}^1/_2[k_o^2+k_e^2\pm(k_o^2-k_e^2)\,(1+A^2)^{1/2}]. \quad (22)$$

We shall now consider the limiting cases. In the case of moderately small angle $\varphi\sim 1$ the difference between $k_0(\varphi)$ and $k_e(\varphi)$ or between $(\hat\eta_0)_{yy}$ and $(\hat\eta_0)_{vv}$ is of the first order in μ. We then have $A^2\propto\mu^2$, the product $A^2(k_o^2-k_e^2)$ is of the order of μ^3, and the roots of Eq. (22) are

$$k_1^2=k_o^2(\varphi)+O(\mu^3); \quad k_2^2=k_e^2(\varphi)+O(\mu^3). \quad (23)$$

The wave at $|\mathbf{k}|=k_1$ has mainly the polarization $\mathbf{e}_y$, and the wave $|\mathbf{k}|=k_2$ the polarization $\mathbf{e}_v$. If $\varphi=0$, i.e., in the case of propagation exactly along the helix axis we find that

$$(\eta_0)_{yy}=(\eta_0)_{vv}, \quad k_o(\varphi=0)=k_e(\varphi=0),$$

and the roots of Eq. (19) are

$$(k^2)_{1,2}=k_o^2(\varphi=0)\pm\varepsilon^2p^2|\psi_{vy}(\varphi=0)|. \quad (24)$$

The limiting case $|\mathbf{q}|\to\infty$ will be considered below in Sec. 5.

We shall now write down the explicit formulas for cholesterics. In this case, we have

$$\theta=0, \quad \varepsilon_1=\varepsilon_\parallel, \quad \varepsilon_2=\varepsilon_3=\varepsilon_\perp, \quad \varepsilon_\parallel-\varepsilon_\perp=\varepsilon_a$$

and we can obtain

$$\begin{aligned}
(\eta_0)_{yy}&=\frac{\varepsilon_\perp+{}^1/_2\varepsilon_a}{\varepsilon_\perp(\varepsilon_\perp+\varepsilon_a)},\\
(\eta_0)_{vv}&=\frac{\varepsilon_\perp+{}^1/_2\varepsilon_a}{\varepsilon_\perp(\varepsilon_\perp+\varepsilon_a)}\cos^2\varphi+\frac{1}{\varepsilon_\perp}\sin^2\varphi,\\
\psi_{yy}&=-\left(\frac{\varepsilon_a}{\varepsilon}\right)^2\frac{p^2\varepsilon(1-7\cos^2\varphi)+8q^2}{32\varepsilon(q^2-p^2\varepsilon\cos^2\varphi)},\\
\psi_{vv}&=\psi_{yy}\cos^2\varphi, \quad \psi_{yv}^*=\psi_{vy}=-i\left(\frac{\varepsilon_a}{\varepsilon}\right)^2\frac{p^3\varepsilon^{1/2}\cos^2\varphi(1+\cos^2\varphi)}{32q(q^2-p^2\varepsilon\cos^2\varphi)}.
\end{aligned} \quad (25)$$

In the case of relatively small angles the wave is of the o type, characterized by $\mathbf{D}\sim\mathbf{e}_y$ and by a small ellipticity:

$$\frac{D_y}{D_v}=im=i\varepsilon_a\frac{p^3\varepsilon^{1/2}(1+\cos^2\varphi)\operatorname{ctg}^2\varphi}{16q(q^2-p^2\varepsilon\cos^2\varphi)}. \quad (26)$$

In the case of an e-type wave, the ellipticity is also small: $D_v/D_y=im$.

The transition from the regime of linear ($\varphi\sim 1$) to the regime of circular ($\varphi=0$) birefringence occurs at $\varphi\sim(\varepsilon_a/\varepsilon)^{1/2}$. In this region we can make the substitution $\sin^2\varphi\to\varphi^2$ in all the expressions and, moreover, we can regard φ^2 as a small parameter proportional to ε_a. With the same precision, we obtain

$$\begin{aligned}
(k^2)_{1,2}&=p^2\varepsilon_\perp\left(1+\frac{\varepsilon_a}{2\varepsilon_\perp}-\frac{1}{4}\frac{\varepsilon_a^2}{\varepsilon_\perp^2}-\frac{1}{4}\frac{\varepsilon_a\varphi^2}{\varepsilon_\perp}\right)\\
&\pm\frac{p^2\varepsilon_a}{4}\left[\varphi^4+\frac{\varepsilon_a^2\varepsilon p^6}{16q^2(q^2-p^2\varepsilon)^2}\right]^{1/2}+p^2\varepsilon\left(\frac{\varepsilon_a}{\varepsilon}\right)^2\frac{4q^2-3p^2\varepsilon}{16(q^2-p^2\varepsilon)},\\
\frac{D_y}{D_v}&=i[x\pm(1+x^2)^{1/2}], \quad x=\frac{\varphi^2}{\varepsilon_a}\frac{4q(q^2-p^2\varepsilon)}{p^3\varepsilon^{1/2}}.
\end{aligned} \quad (27)$$

We can see from Eq. (27) that the parameter x describing the transition from the circularly to the linearly polarized normal waves contains not only the factor $\varepsilon\varphi^2/\varepsilon_a$ but also the dimensionless factor $q(q^2-k^2)/k^3$. If $q\sim k$, there is no resonance and this second factor is of the order of unity. On approach to a resonance the role of the virtual waves increases and the range of circular normal waves, characterized by $0\leqslant\varphi\leqslant x$ and $x\sim 1$, becomes wider. In the limit $q\to 0$, this range again widens formally, but the situation is no longer described by our results because in the limit $q\to 0$ the adiabatic Mauguin limit is reached.

5. QUASISTATIC APPROXIMATION

In the case when the period of a helix is much less than the wavelength of light in the liquid-crystal medium, i.e., when $|\mathbf{q}|\gg p\varepsilon^{1/2}$, the electric field can be regarded as quasistatic over one pitch of the helix, i.e., the Maxwell equations can be replaced with

$$D_i(\mathbf{r})=\varepsilon_{ik}(\mathbf{r})E_k(\mathbf{r}), \quad (28a)$$

$$\operatorname{div}\mathbf{D}(r)=0, \quad \operatorname{rot}\mathbf{E}(\mathbf{r})=0. \quad (28b)$$

We shall assume that $\varepsilon_{ik}(\mathbf{r})$, $\mathbf{E}(\mathbf{r})$, and $\mathbf{D}(\mathbf{r})$ can be represented in the form of spatially averaged parts and fluctuations with zero mean value:

$$\varepsilon_{ik}(\mathbf{r})=\bar\varepsilon_{ik}+\delta\varepsilon_{ik}(\mathbf{r}), \quad \mathbf{D}(\mathbf{r})=\bar{\mathbf{D}}+\delta\mathbf{D}(\mathbf{r}), \quad \mathbf{E}(\mathbf{r})=\bar{\mathbf{E}}+\delta\mathbf{E}(\mathbf{r}). \quad (29)$$

We then have the problem of calculation of the effective permittivity of such a medium, i.e., of the coefficient $\varepsilon_{ik}^{\text{eff}}$ in the relationship

$$\bar D_i=\varepsilon_{ik}^{\text{eff}}\bar E_k. \quad (30)$$

In the case when the fluctuations $\delta\varepsilon_{ik}(\mathbf{r})$ are, firstly, scalar, i.e.,

$$\delta\varepsilon_{ik}(\mathbf{r})=\delta\varepsilon(\mathbf{r})\delta_{ik},$$

and, secondly, are isotropically correlated

$$\langle\delta\varepsilon(\mathbf{r}_1)\delta\varepsilon(\mathbf{r}_2)\rangle=f(|\mathbf{r}_1-\mathbf{r}_2|),$$

this problem was solved in Ref. 8 (§9), where the permittivity of a mixture was calcualted. We shall show how to solve this problem without recourse to the above restrictions.

We shall adopt the Fourier representation

$$A(\mathbf{r})=\bar A+\sum_{\mathbf{k}}e^{i\mathbf{kr}}\tilde A(\mathbf{k}), \quad (31)$$

where A is any of the quantities in Eq. (29) and $\tilde A(\mathbf{k})$ does not contain the term with $\mathbf{k}=0$. Then, with precision quadratic in respect of $\delta\varepsilon$, we have

$$\tilde D_i(\mathbf{k})=\bar\varepsilon_{ik}\tilde E_k(\mathbf{k})+\tilde\varepsilon_{ik}(\mathbf{k})\bar E_k, \quad (32)$$

$$\bar D_i=\bar\varepsilon_{ik}\bar E_k+\sum_{\mathbf{k}}\tilde\varepsilon_{ij}(\mathbf{k})\tilde E_j(-\mathbf{k}). \quad (33)$$

The vector equation (32) (i.e., three relationships) links six unknowns: $\tilde E_x$, $\tilde E_y$, $\tilde E_z$, $\tilde D_x$, $\tilde D_y$, and $\tilde D_z$. Therefore, for the purpose of the single-value determination of these unknowns in terms of the known quantities ($\bar{\mathbf{E}}$, $\bar\varepsilon_{ik}$, and $\tilde\varepsilon_{ik}$) it is necessary to use the electrostatic equations of the system (28b). They can be solved particularly simply in the Fourier

representation:

$$\bar{E}_i(\mathbf{k}) = (k_i/k)\bar{E}(\mathbf{k}), \quad (\tilde{\mathbf{D}}(\mathbf{k})\mathbf{k}) = 0, \tag{34}$$

i.e., the vector $\bar{\mathbf{E}}$ is purely longitudinal and has just one independent component, whereas the vector $\mathbf{D}$ is purely transverse (with respect to $\mathbf{k}$) and has two independent components. For these quantities the system (32) is now fully determinate. In particular, combining Eq. (32) with the vector $\mathbf{k}$, we obtain

$$\bar{E}_j(\mathbf{k}) = -k_j k_m \tilde{\varepsilon}_{mk}(\mathbf{k})\bar{E}_k/\bar{\varepsilon}_{en}k_e k_n. \tag{35}$$

Substituting this expression in Eq. (33), we find that the effective permittivity is given by

$$\varepsilon_{ik}^{\text{eff}} = \bar{\varepsilon}_{ik} - \sum_{\mathbf{k}} \frac{k_j k_m \tilde{\varepsilon}_{ij}(\mathbf{k})\tilde{\varepsilon}_{mk}(-\mathbf{k})}{\bar{\varepsilon}_{en}k_e k_n}. \tag{36}$$

In the specific case of fluctuations the expression on the right-hand side of Eq. (36) should be averaged over an ensemble and then, in principle, it can be expressed in terms of the spatial correlation function $\langle \delta\varepsilon_{ik}(\mathbf{r}_1)\delta\varepsilon_{em}(\mathbf{r}_2)\rangle$. In the case of scalar fluctuations if $\bar{\varepsilon}_{ik} = \bar{\varepsilon}\delta_{ik}$ and in the situation when the correlation function $\langle \tilde{\varepsilon}(\mathbf{k})\tilde{\varepsilon}(-\mathbf{k})\rangle$ depends only on $|\mathbf{k}|$, the tensor $k_j k_m/k^2$ may be replaced with $\delta_{jm}/3$ and then Eq. (36) yields the well known result (see §9 in Ref. 8):

$$\varepsilon^{\text{eff}} = \bar{\varepsilon} - {}^1/_3\delta\varepsilon^2/\bar{\varepsilon}. \tag{37}$$

We shall now apply Eq. (36) to the problem of a chiral liquid crystal with $|\mathbf{q}| \gg p\varepsilon^{1/2} = \omega\varepsilon^{1/2}/c$. The substitution of Eqs. (2) and (3) in Eq. (36) gives

$$\varepsilon_{xx}^{\text{eff}} = \varepsilon_{yy}^{\text{eff}} = (\varepsilon_0)_{xx} - \frac{1}{8}\frac{(\varepsilon_2-\varepsilon_3)^2}{(\varepsilon_0)_{zz}}\sin^2 2\theta, \tag{38}$$

$$\varepsilon_{xy} = \varepsilon_{xz} = \varepsilon_{yz} = 0, \quad \varepsilon_{zz}^{\text{eff}} = (\varepsilon_0)_{zz}.$$

In particular, in the case of cholesterics we have $\theta = 0$ and $\varepsilon_2 = \varepsilon_3$, so that if $|\mathbf{q}| \gg p\varepsilon^{1/2}$, then

$$\varepsilon_{ik} = \bar{\varepsilon}_{ik} + O(\varepsilon_a^3). \tag{39}$$

It follows that in the limit $q \to \infty$ it is possible to deal with cholesterics by an approach in which the tensor $\hat{\varepsilon}$ and not the tensor ε^{-1} is averaged in space. In the case of smectics, none of the above approaches is applicable and only an expression of the (36) type is correct. Finally, if $|\mathbf{q}| \sim p\varepsilon^{1/2}$ (but not near a Bragg resonance) the complete electrodynamic problem can be solved.

It is interesting to study also the behavior of the specific rotation of the plane of polarization $d\alpha/dz$ in the course of propagation along the z axis. In this case the specific rotation for cholesterics is

$$\frac{d\alpha}{dz} = \left(\frac{\varepsilon_a}{\varepsilon}\right)^2 \frac{k^4}{32q(q^2-k^2)}; \quad k^2 = p^2\varepsilon, \tag{40}$$

whereas in the case of smectics, it is

$$\frac{d\alpha}{dz} = \frac{k^4}{32q(q^2-k^2)}(\mu_1 - \mu_3\sin^2\theta)^2. \tag{41}$$

A characteristic feature is the fast (as q^{-3}) fall of the gyrotropy in the limit $q \to \infty$. It therefore follows that the gyrotropy is in this case an effect of the third order of smallness with respect to a/λ, where $a \propto q^{-1}$ is the pitch of the helix.

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[1]C. W. Oseen, Trans. Faraday Soc. **29**, 883 (1933).
[2]H. de Vries, Acta Crystallogr. **4**, 219 (1951).
[3]E. I. Kats, Zh. Eksp. Teor. Fiz. **59**, 1854 (1970) [Sov. Phys. JETP **32**, 1004 (1971)].
[4]V. A. Belyakov and V. D. Dmitrienko, Fiz. Tverd. Tela (Leningrad) **15**, 2724 (1973) [Sov. Phys. Solid State **15**, 1811 (1974)].
[5]P. G. de Gennes, The Physics of Liquid Crystals, Clarendon Press, Oxford, 1974 (Russ. Transl., Mir, M., 1977).
[6]L. N. Blinov, Élektro-i magnitooptika zhidkikh kristallov (Electro- and Magnetooptics of Liquid Crystals), Nauka, M., 1978).
[7]V. A. Belyakov, V. D. Dmitrienko, and V. P. Orlov, Usp. Fiz.Nauk **127**, 221 (1979) [Sov. Phys. Usp. **22**, 63 (1979)].
[8]L. D. Landau and E. M. Lifshitz, Élektrodinamika sploshnvkh sred, Gostekhizdat, M., 1957 (Electrodynamics of Continuous Media, Pergamon Press, Oxford, 1960).

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