Optical activity in the isotropic phase of cholesteric liquid crystals

E. I. Kats

L. D. Landau Institute of Theoretical Physics, USSR Academy of Sciences
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The fluctuation rotation of the plane of polarization of transmitted light in cholesteric liquid crystals above the transition point (T_c) is considered in the self-consistent-field approximation. It is shown that the sign of the optical activity does not depend on the incident-light wavelength (there is no inversion of the optical activity). An analogous investigation is made of the effect of fluctuations on the rotation of the polarization plane at $T < T_c$. The results agree with the experimental data.

1. INTRODUCTION

Cholesteric liquid crystals (CLC) are made up of molecules of elongated form, having no inversion center. Therefore the ground state of a CLC is inhomogeneous. By virtue of this inhomogeneity, the CLC have rather unique optical properties, and in particular a tremendous rotation of the plane of polarization (φ). The value of φ is connected with the inhomogeneity of the ordering of the molecules and greatly exceeds the molecular rotation of the plane of polarization (φ_0) due to the spatial dispersion of the dielectric tensor. The transition of a CLC from the isotropic phase to the ordered phase is a first-order transition. In most liquid crystals, however, this transition is very close to a second-order transition (the coefficient of the cubic invariant in the Landau expansion of the free energy is anomalously small). It is therefore natural to expect fluctuating pre-transition phenomena to appear, even in the isotropic phase, as the transition temperature is approached. Particular interest attaches to the optical activity that is typical of CLC only (whereas the fluctuation increase of the light scattering occurs in the isotropic phase of nematic liquid crystals). It should be borne in mind, however, that a molecular optical activity φ_0 is also present in the isotropic phase. Therefore to observe the fluctuations of the optical activity it is necessary to have $\varphi > \varphi_0$. As will be shown below, this usually occurs at the parameter values typical of CLC in a wide range of temperatures (~10°) near the transition point. In addition, φ_0 is practically independent of the temperature, so that the fast temperature growth of φ can be separated against this background. Since a real first-order transition takes place at a temperature T_c that differs little from the temperature T_c^* of the fictitious second-order transition, the increase of φ if "cut off" at the temperature T_c.

The optical properties of the ordered phase of CLC were considered by several workers (see, e.g., [1]). It is possible in this case to obtain an exact solution of Maxwell's equations, owing to the simple exponential dependence of the dielectric-tensor components on the coordinates ($\exp(\pm 2i\alpha z)$, where $2\pi/\alpha$ is the period of the helical structure). In the isotropic phase, on the other hand, the fluctuation corrections to the dielectric tensor have a continuous spectrum of harmonics with a maximum at the wave vector 2α. In Sec. 2 we therefore derive formulas for the optical activity in a medium with small and random inhomogeneities. The derivation is carried out with the aid of a diagram technique for the Green's function of the radiation in the medium. This is equivalent to the use of the methods of nonlinear oscillation theory. The angle of rotation of the plane of polarization in CLC depends nonlinearly on the path traversed by the light in the medium. It is therefore more convenient to calculate the differential optical activity $\Delta k = \partial\varphi/\partial z$ at $z = 0$. At small thicknesses of the CLC we have $\varphi \approx l\Delta k$. In the isotropic phase, Δk is expressed by an integral of the correlation function of the dielectric-tensor components. The correlators necessary for the calculation of Δk are calculated in Sec. 3 in the self-consistent-field approximation. As the temperature approaches T_c^*, the value of Δk increases like $(T - T_c^*)^{-1/2}$, in agreement with recent experiments [2]. In addition, Δk has a constant sign, whereas in the ordered phase of the CLC, at an optical wavelength coinciding with the period of the structure, the sign of the optical activity changed.

Section 4 is devoted to fluctuations at $T < T_c$. Lubensky [3] has shown that CLC are similar, in the sense of their symmetry, to two-dimensional degenerate systems (Bose liquid, Heisenberg magnets, etc.). The fluctuations of the order parameter therefore diverge, just as in the two-dimensional case. In CLC this is manifest in the fact that the correlation function of the order parameter is of the form

$$C = T/(k_3^2 + \xi^2 k_\perp^4), \qquad (1)$$

k_3 is the wave-vector component along the helix axis (z); $\mathbf{k}_\perp$ is the wave-vector component in a plane perpendicular to the axis; ξ is a constant $\sim 1/\alpha$. If $\mathbf{m}$ were a unit vector in the direction of the helix axis, then the ordered phase of the CLC would be specified by the condition $\mathbf{m} = \text{const}$. The fact that the integral of C, as given by formula (1), diverges logarithmically means that $\langle \mathbf{m} \rangle = 0$ at all finite temperatures. In analogy with two-dimensional systems [4], however, a phase transition takes place nevertheless, but the true order parameter is not the "spontaneous moment" $\mathbf{m}$, but a property analogous to the transverse rigidity of magnets. In CLC there is a special small quantity ($\alpha a \ll 1$, where a is the atomic dimension), which makes the low-temperature phase almost ordered down to T_c. With the aid of the formulas of Sec. 2 we can calculate the optical activity in this case too. Small corrections ($\sim \alpha a$) to the formulas for the polarization-plane rotation of the ordered phase of the CLC are obtained. Fluctuations in the transition region near T_c are also discussed.

2. OPTICAL ACTIVITY AT $T > T_c$

Let us consider an electromagnetic wave propagating in the z-axis direction in the isotropic CLC phase. The electric field in the medium therefore has components E_x and E_y. Since the rotation of the polarization plane is the difference between the propagation velocities of left- and right-polarized waves, it is convenient to introduce the circular components $E_\pm = E_x \pm iE_y$. The isotropic phase is characterized by a dielectric tensor $\epsilon_{ij} = \epsilon_0 \delta_{ij}$ (δ_{ij} is a unit tensor), and

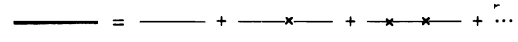

FIG. 1

in this case only the molecular optical activity φ_0 is present. The polarization-plane rotation connected with the inhomogeneity arises only when account is taken of the fluctuation corrections to the tensor ϵ_{ij}. When light propagates along the z axis, only the components $\chi_\pm = \epsilon_{11} \pm i\epsilon_{12}$ are of importance. If we introduce the retarded Green's function D_{ij}^R of the radiation in the usual manner [5], then, without allowance for the fluctuations, this function is given by

$$D_{ij}^R = \frac{4\pi}{\varepsilon_0\omega^2/c^2 - k^2}\left(\delta_{ij} - \frac{k_i k_j}{k^2}\right) \tag{2}$$

(in a gauge where $\operatorname{div}\mathbf{A} = 0$ and $\mathbf{A}$ is the vector potential). The optical activity means that the poles of the Green's functions D_{ik}^E of Maxwell's equations for the right- and left-hand polarizations are unequal. Then [5]

$$D_{ik}^E = \omega^2 D_{ik}^R. \tag{3}$$

In the case when $\epsilon_{ik} = \epsilon_0\delta ik$, as follows from (2), there is no optical activity. However, owing to the fluctuations there is the interaction

$$H_{int} = \omega^2 \int \chi_{ik} A_i A_k \, dv. \tag{4}$$

Dyson's equation for the Green's function $\tilde{D}_{ik}^R$ with allowance for the interaction (4) is shown in Fig. 1 (the cross on the diagram indicates the interaction (4)). By virtue of the random character of the fluctuations we have $\langle\chi_\pm\rangle = 0$ but $\langle\chi_\pm\chi_\mp\rangle \neq 0$. Therefore the equation for $\tilde{D}_{ik}^R$ should be averaged in analogy with the procedure used when random impurities are taken into account. Unlike the impurity problem, however, the averaging if carried out over a Gibbs ensemble and not over the impurity positions. As a result, we obtain

$$\tilde{D}_{ik}^R = \frac{4\pi}{k_0^2 - q^2 - D}\left(\delta_{ik} - \frac{q_i q_k}{q^2}\right), \tag{5}$$

where $k_0 = \sqrt{\epsilon\omega}/c$ is the wave vector in the medium and

$$D = \frac{\delta^2 k_0^3}{2} \int \langle \chi_{im}\chi_{mk}\rangle \, \tilde{D}_{ik}^R \, d^3q. \tag{6}$$

Formula (6) thus gives the frequency renormalization due to the influence of the fluctuations. From this we can obtain the differential optical activity directly. In second order in the fluctuations, the corresponding diagram is shown in Fig. 2 (the wavy line denotes the fluctuation propagator $\langle\chi_+\chi_-\rangle$):

$$\Delta k = \frac{\delta^2 k_0^3}{2} \int d^3q \left\{ \frac{\langle \chi_+(\mathbf{q}-\mathbf{k}_0)\chi_-(-\mathbf{q}+\mathbf{k}_0)\rangle}{k_0^2 - q^2} - \frac{\langle \chi_+(\mathbf{q}+\mathbf{k}_0)\chi_-(-\mathbf{q}-\mathbf{k}_0)\rangle}{k_0^2 - q^2} \right\},$$

If we put in the first term of this expression $\mathbf{q} = \mathbf{k}_0 \equiv \kappa$ and in the second $\mathbf{q} + \mathbf{k}_0 \equiv \kappa$, then we obtain

$$\Delta k = \delta^2 k_0^3 \int d^3\varkappa \frac{\langle \chi_+(\varkappa)\chi_-(-\varkappa)\rangle (2\varkappa_z k_0 + \varkappa_\perp^2)}{(2\varkappa_z k_0 + \varkappa_\perp^2)^2 - \varkappa_z^4}. \tag{7}$$

The same formula can be used to calculate the optical activity in the ordered phase ($T < T_c^*$). In this case there is no longer any need for averaging, since we have a nonzero $\langle\chi_\pm\rangle = \exp(\pm 2i\alpha z)$. Therefore, integrating (7), we obtain

$$\Delta k = \delta^2 k_0^4 / 4\alpha(k_0^2 - \alpha^2). \tag{7'}$$

Formula (7') coincides with the results of the exact solution of Maxwell's equations in this case [1].

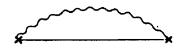

FIG. 2

For comparison, the Appendix contains a derivation of formula (7) based directly on a solution of Maxwell's equations in a random medium. We note also that formula (7), which expresses the optical activity due to the short-range order $\langle\chi_+\chi_-\rangle$, can have a wider range of applicability. As seen directly from the derivation, it is applicable to any system with weak inhomogeneity and anisotropy. Cheng and Meyer [2] calculated the optical activity of CLC by using the concept of rotation of the polarization plane in a system of inactive groups coupled by dipole-dipole forces. This approach cannot describe the CLC adequately, first because their components themselves have optical activity and second, dipole-dipole forces alone cannot ensure stability of the CLC in any case.

3. CORRELATIONS IN THE ISOTROPIC PHASE OF CLC

We write down the most general form of the free energy compatible with the symmetry of the CLC. It is more convenient here to expand not in the gradients of the deviation of the orientation from equilibrium, but in terms of physical quantities of interest to us, namely the anisotropic parts of the dielectric tensor. In analogy with [2], we have

$$\delta F = {}^1/_2 A\varepsilon_{ij}^2 + {}^1/_2 C(\nabla\varepsilon_{ij})^2 + \alpha C e_{ijk}\varepsilon_{im}\nabla_k\varepsilon_{jm}. \tag{8}$$

The third term takes into account the absence of an inversion center in CLC (α is a pseudoscalar quantity), and all the phenomena of interest to us are connected with the presence of this term; A and C are coefficients of the Landau expansion ($C = \text{const} > 0$, $A \sim \sqrt{\tau}$, $\tau = T - T_c^*$). In formula (8) we have left out the fourth-order terms, which are not of interest to us, and for simplicity we have disregarded the presence of the second correlation length.

As already noted in Sec. 2, an important role in the calculation of the optical activity is played by the z-dependent components of the tensor ϵ_{ij}. This singles out immediately the corresponding two-dimensional representation for the free energy (8)

$$\varepsilon_{ij} = \begin{pmatrix} \varepsilon_{11} & \varepsilon_{12} \\ \varepsilon_{21} & \varepsilon_{22} \end{pmatrix}. \tag{9}$$

The general requirement that the tensor ϵ_{ij} be symmetrical yields the condition $\epsilon_{12} = \epsilon_{21}$. In addition, in CLC the order parameter has a zero trace, and this calls for $\epsilon_{11} = -\epsilon_{22}$. At $T < T_c^*$ [1] we have

$$\varepsilon_{11} = \delta\varepsilon \cos 2\alpha z, \quad \varepsilon_{12} = \delta\varepsilon \sin 2\alpha z$$

(ϵ is the average dielectric constant). It is therefore convenient to choose the following two quantities to characterize the order parameter:

$$\varepsilon_{11} = -\varepsilon_{22} = S\cos\theta, \quad \varepsilon_{21} = \varepsilon_{12} = S\sin\theta. \tag{10}$$

Substituting (10) in (8) we obtain

$$\delta F = AS^2 + C(\nabla S)^2 + CS^2[(\nabla\theta)^2 - 2\alpha\nabla_3\theta]. \tag{11}$$

Formula (11) corresponds to a transition to the ordered state of CLC in the form of a spreading of the fluctuation regions with helical order. The dimension of each such region $\sim(C/A)^{1/2}$ becomes infinite at the transition

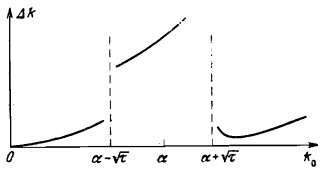

FIG. 3

point. Of course, all this is true only in the first-order self-consistent-field approximation. As already noted in the Introduction, when a certain proximity to T_c is reached, both the interaction of the fluctuations and the presence of helix-axis orientation fluctuations, which destroy the helical order, become important (see Sec. 4 for details).

It follows from (11) that the correlation function $\langle\chi_+\chi_-\rangle$ of interest to us can be expressed in terms of the correlator S. Indeed,

$$\chi_+(\varkappa)=\varepsilon_{11}(\varkappa)+i\varepsilon_{12}(\varkappa)=S(\varkappa_z+2\alpha,\ \varkappa_\perp),$$
$$\chi_-(-\varkappa)=\varepsilon_{11}(-\varkappa)-i\varepsilon_{12}(-\varkappa)=S(-\varkappa_z-2\alpha,\ -\varkappa_\perp). \tag{12}$$

Carrying out simple integration (averaging with the free energy (8)), we obtain

$$\langle\chi_+(\varkappa)\chi_-(-\varkappa)\rangle=T/[\tau+(\varkappa_z+2\alpha)^2+\varkappa_\perp^2]. \tag{13}$$

We have taken into account here the fact that the true transition temperature is determined by the vanishing of the renormalized coefficient of S^2. This corresponds to separation of the complete square in (11) and to $\tau=(A/C)+4\alpha^2$.

Substituting (13) in (11) we have

$$\Delta k=2\delta^2Tk_0^4\int\frac{d^3\varkappa\,(2k_0\varkappa_z+\varkappa_\perp^2)}{[\tau+(\varkappa_z+2\alpha)^2+\varkappa_\perp^2][(2k_0\varkappa_z+\varkappa_\perp^2)^2-\varkappa_z^4]}. \tag{14}$$

The value of the integral in (14) depends on the ratio of $k_0-\alpha$ and $\sqrt{\tau}$. In the case when $|k_0-\alpha|<\sqrt{\tau}$ we have

$$\Delta k=\tfrac{1}{8}\delta^2Tk_0^2\alpha/\sqrt{\tau} \tag{15}$$

(the nonsingular terms of the type $\tau\ln\alpha\tau^{-1}$ have been left out).

On the other hand, if $|k_0-\alpha|\gg\sqrt{\tau}$, then the principal contribution to the integral is made by the integration region $(\varkappa_z+2\alpha)^2>\varkappa_\perp^2$. Then Δk is small and strong scattering obtains:

$$\Delta k=\frac{\delta^2T}{16}\frac{k_0^2\tau}{(k_0-\alpha)^2}. \tag{16}$$

The dependence of Δk on the wavelength at a given temperature (and consequently at a given τ) is shown schematically in Fig. 3. The temperature dependence of Δk agrees with the experimental data [2].

4. FLUCTUATIONS OF THE ORDER PARAMETER AT $T<T_c^*$

As already indicated in Introduction, helical ordering of CLC is impossible at any finite temperature. This is reflected in the form of the correlation function C (see formula (1)). The nature of the low-temperature phase $T<T_c^*$ becomes manifest in the power-law fall-off the fluctuations. This circumstance can be discerned directly by writing down the Euler-Lagrange equations for the functional (8). The roots of the dispersion equation then satisfy the condition $k_z\sim k_\perp^2$. This indeed conforms to formula (1). The Hamiltonian of the fluctuations is obtained from the Franck expansion of the free energy for CLC

$$H-E_0=K\int d^3x\{(\nabla_3\varphi)^2+\xi^2(\nabla_\perp^2\varphi)^2\}. \tag{17}$$

Here φ is a parameter characterizing the helical order (the polar angle of the director) and K is the modulus of elasticity.

Instead of (17) we can write down a fully two-dimensional expression. To this end we introduce a new field Φ:

$$\int\xi^2(\nabla_\perp^2\varphi)^2\,dx_3\equiv\xi(\nabla_\perp\Phi)^2$$

(ξ has been introduced to preserve dimensionality in the right-hand side). We next integrate the first term in the right-hand side of (17) by parts. The integral term vanishes by virtue of symmetry, and we incorporate the remaining expression in E_0. This represents the nonsingular fluctuations in a plane perpendicular to $\mathbf{m}$. We then get from (17)

$$H-E_0=J\int d^2x(\nabla_\perp\Phi)^2. \tag{18}$$

Thus, in the long-wave approximation the CLC are indeed equivalent to two-dimensional degenerate systems. The wave vector along the z axis can be arbitrary in the interval $[-\alpha,\ \alpha]$.

In formula (18) we have put, in order of magnitude, $K=T_c/a$ (a is the atomic dimension). Therefore $J=T_c\xi/2\pi a$, and since $\xi\sim1/\alpha$, the low-temperature condition $T\ll J$ is always satisfied in real CLC. This means that CLC are always "almost" ordered. Since $\alpha a\sim10^{-2}$ to 10^{-3}, divergences would occur at astronomical distances $\sim10^{50}$ cm, meaning that the destruction of long-range order would take place at astronomical times. In the low-temperature region we can obtain from (1)

$$\langle\cos\theta\cos\theta'\rangle=\text{const}\cdot\cos\alpha(z-z')R^{-\gamma}. \tag{19}$$

Here

$$\gamma=\frac{T}{2\pi K\xi}=\frac{Ta}{2\pi T_c\xi},\qquad R=\frac{|z|}{\xi}+\frac{r_\perp^2}{\xi^2}.$$

With the aid of (19) and (7) we obtain

$$\Delta k=(1-\gamma)\delta^2k_0^4/4\alpha(k_0^2-\alpha^2). \tag{20}$$

Since $\gamma\ll1$, formula (20) naturally differs little from formula (7′) corresponding to the fully-ordered state. Relation (19) indicates, however, that the polarization-plane rotation angle is nonlinear even in small layers of CLC, $\varphi\sim l^{1-\gamma}$. We point out that the entire derivation of formulas (19) and (20) is connected with the use of the Franck expansion. The divergences are the result of the fact that the terms $\sim q_\perp^2$ cancel out in the lowest mode of the CLC. This, of course, is no accident, and reflects the symmetry of the CLC. In analogy with the derivations of Lubensky [3], we can demonstrate that when the next higher terms of the expansion of the free energy in the deviations from equilibrium are taken into account, $q_\perp^2$ is cancelled out as before. To this end it is necessary only to have a sufficient short-range action, namely, the effective radius of the forces should be much shorter than the period of the produced structure. But if, for example, the van der Waals forces are taken into account, terms $\sim q_\perp^3$ are added to the Franck free energy, and there are no divergences. Of course, the van der Waals forces can stabilize the CLC only if the cubic term is significant at the same values of the

wave vectors at which the divergences come into play ($q_\perp \sim \alpha$).

We note in conclusion one more circumstance. At $T > T_c^*$ the self-consistent-field approximation is violated in the immediate vicinity of T_c^* (provided that no first-order transition has taken place earlier). It follows from (1) that this violation takes place at

$$\gamma \ln (a\sqrt{\tau}) \sim 1. \tag{21}$$

The presence of fluctuations makes the very transition with order parameter $\langle \mathbf{m} \rangle$ impossible. The self-consistent-field approximation "knows nothing" of this. Therefore the character of the correlation functions should change in the region (21) near the temperature T_c^*. Instead of formula (13) we should have an expression in which the mode does not "soften" at $\kappa_z = -2\alpha$. This can be taken into account qualitatively with the aid of the following artificial strategem. Formula (13) is the result of choosing the order parameter in the form (10) corresponding to the ordered phase of the CLC. The fact that such a phase is impossible means that in the vicinity (21) of T_c^* the order parameter is of a different form. We choose in place of (10) the representation

$$\hat{\varepsilon} = \begin{pmatrix} \varphi_1 & \varphi_2 \\ \varphi_2 & -\varphi_1 \end{pmatrix}. \tag{22}$$

Here φ_1 and φ_2 are already independent quantities (unlike in (10), where $\varphi_1^2 + \varphi_2^2 = \text{const}$).

Substituting (22) in (8) and carrying out simple integration, we obtain

$$\langle \varphi_1^2 \rangle = \langle \varphi_2^2 \rangle = T \left[\tau + k_z^2 + k_\perp^2 + \frac{4\alpha^2 k_z^2}{\tau + k_z^2 + k_\perp^2} \right]^{-1}. \tag{23}$$

At $\tau \ll k_Z^2$ and $k_\perp^2 \ll k_Z^2$ we have from (23)

$$\langle \varphi^2 \rangle = T/(k_z^2 + 4\alpha^2). \tag{24}$$

The form of formula (24) (the absence of a real pole) agrees with the fact that there is no transition into the helical state of the CLC.

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APPENDIX

Maxwell's equations take the following form in terms of the components $E_\pm$:

$$\Delta E_\pm + k_0^2 E_\pm + \delta k_0^2 \chi_\pm E_\mp = 0. \tag{A.1}$$

In the zeroth order in the fluctuations, the equations for the circular components of the field separate, and there is no optical activity:

$$E_\pm^{(0)} = A_\pm e^{ik_0 z} + B_\pm e^{-ik_0 z}. \tag{A.2}$$

We seek the solution in first-order perturbation theory in the form $E_\pm = E_\pm^{(0)} + E_\pm^{(1)}$, where the zero-order solution (A.2) has a renormalized frequency $k_\pm = k_0 + k_\pm^{(1)}$. The renormalization is analogous to the condition for the absence of resonance in nonlinear systems [6]

$$k_\pm^{(1)} = 0, \quad E_\pm^{(1)}(\mathbf{q}) = -\delta k_0^2 \left[\frac{A_\mp \chi_\pm(\mathbf{q}-\mathbf{k}_0)}{k_0^2 - q^2} - \frac{B_\mp \chi_\pm(\mathbf{q}+\mathbf{k}_0)}{k_0^2 - q^2} \right]. \tag{A.3}$$

Here $E_\pm^{(1)}(\mathbf{q})$ and $\chi_\pm(\mathbf{q})$ are the Fourier components of the corresponding quantities. It is seen from (A.3) that, as expected, there is no optical activity in first order in the fluctuations. The frequency renormalization comes into play only in the quadratic expansion in terms of small inhomogeneities

$$\begin{aligned} k_\pm &= k_0 + k_\pm^{(2)}, \\ \Delta E_\pm^{(2)} + k_0^2 E_\pm^{(2)} &= 2k_0 k_\pm^{(2)} E_\pm^{(0)} - k_0^2 \delta \chi_\pm E_\mp^{(1)}. \end{aligned} \tag{A.4}$$

Hence, changing over to Fourier components, we have

$$k_+^{(2)} = -\frac{\delta^2 k_0^3}{2} \int \frac{\chi_+(\mathbf{k}_0 - \mathbf{q}) \chi_-(\mathbf{q} - \mathbf{k}_0)}{k_0^2 - q^2} d^3q. \tag{A.5}$$

Making a change of the integration variable, $\kappa \equiv (k_0 - q_3, \mathbf{q}_\perp)$, we obtain

$$k_+^{(2)} = \frac{\delta^2 k_0^3}{2} \int \frac{\chi_+(\varkappa)\chi_-(-\varkappa)}{\varkappa_z(2k_0 - \varkappa_z) + \varkappa_\perp^2} d^3\varkappa \tag{A.6}$$

and analogously

$$k_-^{(2)} = -\frac{\delta^2 k_0^3}{2} \int \frac{\chi_+(\varkappa)\chi_-(-\varkappa)}{\varkappa_z(2k_0 + \varkappa_z) + \varkappa_\perp^2} d^3\varkappa. \tag{A.7}$$

Therefore the phase difference, which is the cause of the differential rotation of the polarization plane, takes a form that coincides with formula (7) of the text.

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