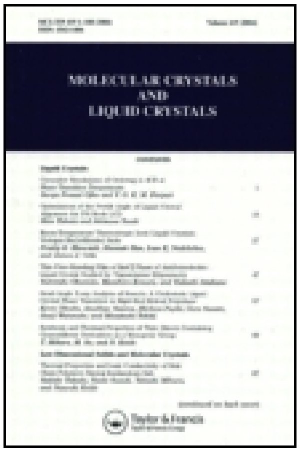

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ON THE APPROXIMATE METHODS FOR LINEAR OPTICS OF LIQUID CRYSTALS

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Abstract Several approximate methods have been proposed so far to treat electromagnetic wave propagation in stratified anisotropic media, such as liquid crystals or aperiodic structures. Despite the complete equivalence among many of them, however, a comparison between different results is often made difficult by the complexity of the adopted formalism. In this paper, an exact propagation equation is given for a plane e.m. wave travelling within a stratified medium. On such a basis, various types of already known approximate results are discussed and generalized in a very simple way.

INTRODUCTION

Since the very discovery of the liquid crystalline state, the attention of many researchers has been addressed to the really spectacular optical properties of some periodic structures [1,2] (cholesterics and, more recently, chiral smectics) which are used in the most important applications of liquid crystals.
Since simple analytic solutions are available only for light travelling along the helix axis [3,4], a great deal of effort has been devoted to develop approximate methods [5-10]. In the last years, the attention of researchers has gradually shifted to the study of methods which can be applied to more general, i.e. aperiodic, structures. The main problem is to

describe the propagation of a plane wave in stratified, anisotropic media, and more-precisely the changes in amplitude and polarization states of a wave propagating in a medium whose dielectric tensor is continuously changing along the direction of a cartesian coordinate, say z. All the existing formal methods introduced to treat light polarization have been used, such as Stokes vectors and Poincaré sphere representation [11-13], Jones vectors [14-16] and different types of generalizations [15-17], Berreman's vectors [18,19] and generalized geometrical optics [20,21]. Some of these methods, and of the obtained results, are fully equivalent. However, the conversion formulae between different methods are generally so complex that it is difficult, except perhaps for a few specialists, to compare the results obtained by different methods. The equivalence, and the conversion expressions between Stokes and Jones vectors have been discussed in some papers [22-24]. A comparison between these two methods and the Bereman vectors method is more complicated, since they are no longer equivalent.

For the above reasons, a review of all these relationships would be of the greatest interest. In the present, paper, a somewhat more restricted, and better defined subject will be dealt with. An exact propagation equation will be given for a plane electromagnetic wave travelling through a stratified medium. In this way, it is particularly easy to generate various types of approximation, (substantially all the methods found in the above quoted papers).

On the basis of this equation, we will discuss various types of already known results and show a simple way to generalize most of them.

For the sake of simplicity we consider a stratified medium whose optical properties can be fully described at any point by a real symmetric dielectric tensor $\underline{\underline{\varepsilon}}$, and assume that $\underline{\underline{\varepsilon}}$ is simply rotating along a generic direction z. An extension to more general cases, such as for instance magnetic, absorbing, or optically active media, is straightforward.

2 - The propagation equation.

We consider a plane wave incident on a slab of an anisotropic medium confined between the planes z = o and z = d of a cartesian coordinate system. The wavevector of the incident light beam is in the plane (x,z), and the dielectric tensor of the anisotropic medium only depend on z. The propagation equation for the electromagnetic wave within the medium can be written in matrix form [18] as:

$$\frac{d\psi}{dz} = i\,k\,\underline{\underline{\Delta}}\,\Psi \qquad (1)$$

where

$$\psi = \begin{pmatrix} E_x \\ H_y \\ E_y \\ -H_x \end{pmatrix}$$

where

$$\underline{\underline{\Delta}} = \begin{pmatrix} \Delta_{11} & \Delta_{12} & \Delta_{13} & 0 \\ \Delta_{21} & \Delta_{11} & \Delta_{23} & 0 \\ 0 & 0 & 0 & 1 \\ \Delta_{23} & \Delta_{13} & \Delta_{43} & 0 \end{pmatrix} \qquad (2)$$

and

$$\Delta_{11} = - m\,\varepsilon_{xz}/\,\varepsilon_{zz}$$

$$\Delta_{12} = 1 - m^2 / \varepsilon_{zz}$$

$$\Delta_{13} = - m\,\varepsilon_{yz}/\,\varepsilon_{zz}$$

$$\Delta_{21} = \varepsilon_{xx} - \varepsilon^2_{xz} / \varepsilon_{zz}$$

$$\Delta_{23} = \varepsilon_{xy} - \varepsilon_{xz}\,\varepsilon_{yz}/\,\varepsilon_{zz} \qquad (3)$$

$$\Delta_{43} = \varepsilon_{yy} - (\varepsilon^2_{yz}/\varepsilon_{zz}) - m^2$$

$$k = \omega/c = 2\pi/\lambda = 1/\bar{\lambda} \;;\; m = k_x/k \qquad (4)$$

If the dielectric tensor is slightly varying over distances of the order of λ, the four differential equations in the variables E_x, H_y, E_y, H_x, can be cast in an equivalent form by choosing a new set of variables which provide two most important advantages:

i) The new variables have the following very simple physical meaning: in any layer dz, they can be considered as the amplitudes a^+, b^+, a^-, b^-, of two forward- and two backward-propagating plane waves.

More precisely, we consider for a given value z_0 of z a fictitious homogeneous medium whose dielectric tensor is $\underline{\underline{\varepsilon}}_0 = \underline{\underline{\varepsilon}}(z_0)$. The waves $\psi_a^+(z)$, $\psi_b^+(z)$, $\psi_a^-(z)$, $\psi_b^-(z)$ are the caracteristic waves of this medium.

The actual function $\psi(z)$ can be expressed, for $z \simeq z_0$, as

$$\psi = a^+ \ \psi_a^+ + b^+ \ \psi_b^+ + \bar{a} \ \psi_a^- - b^- \ \psi_b^- \tag{5}$$

For a locally uniaxial medium $\psi_a^\pm$ and $\psi_b^\pm$ represent the extraordinary and ordinary waves, respectively.

ii) the new differential equations have the form of a set of for coupled equations, with generally small coupling terms, whose meaning is quite clear. For istance the coupling coefficients between ψ_a^+ and ψ_a^- give the relative amplitudes of the a-type reflected waves per unit length along z. Obviously we expect that these coefficients rapidly grow, become no more negligible if the incidence angle is increased beyond the total reflection angle. The considered transformation gives a mathematical basis to physical intuition.

This procedure has been already used in some particular cases [15,16]. Its generalization to the case considered here is straightforward.

We consider the vector transformation

$$\psi(z) = \underline{\underline{T}}(z) \quad \psi_J(z) \tag{6}$$

where $T(z_0)$ is the 4x4 matrix diagonalizing $\underline{\underline{\Delta}}(z_0)$

in $z = z_0$, , i.e. $\underline{\underline{T}}^{-1}\,\underline{\underline{\Delta}}\,\underline{\underline{T}} = \underline{\underline{\Delta}}_D$, where $\underline{\underline{\Delta}}_D$ is diagonal. The components of the new vector $\psi_J(z)$, are the new variables, i.e.

$$\psi_J = \begin{pmatrix} a^+ \\ b^+ \\ a^- \\ b^- \end{pmatrix}, \tag{7}$$

and the propagation equation for ψ_J is

$$\frac{d\psi}{dz} = ik\left[\underline{\underline{\Delta}}_D + i\,\lambda\!\!\!\bar{}\;\underline{\underline{T}}^{-1}\frac{d\underline{\underline{T}}}{dz}\right]\psi_J \tag{8}$$

The coupling coefficients are the elements of the matrix

$$i^2 k\,\frac{2\lambda}{\pi}\,\underline{\underline{T}}^{-1}\,\frac{d\underline{\underline{T}}}{dz} = -\underline{\underline{T}}^{-1}\,\frac{d\underline{\underline{T}}}{dz}$$

For $\underline{\underline{\Delta}}(z)$ = const. the transformation matrix $\underline{\underline{T}}$ is z - independent, and all coupling coefficients are zero.

3 - Approximate solutions.

The coupled equations (8) are a good starting point for the

research of approximate solutions.
Some of the physically most important and used approximations are discussed below.

3.1 Geometrical optics approximation (GOA).

We consider the matrix $i\,\lambda\!\!\!^{-}\, T^{-1}\, dT/dz$ as a perturbation, and expand the function $\Psi_J(z)$ in ascending powers of $\lambda\!\!\!^{-}$:

$$\Psi_J(z) = \sum_{n=o}^{\infty} \lambda\!\!\!^{-\,n}\ \Psi_J^{(n)} \tag{9}$$

The zeroth order solution can be considered the equivalent of the geometrical optics approximations for isotropic media, which corresponds to the limit $\lambda \rightarrow 0$, and of the so called W.K.B. approximation of quantum mechanics.
In this limit, the perturbation term is simply neglected, and the set of equations (8) is decoupled into four independent equations, whose solutions are

$$\begin{aligned} a^{+}(z) &= a^{+}(o)\exp\left[ik\int_{o}^{z}\Delta_a\ (z')\,dz'\right] \\ b^{+}(z) &= b^{+}(o)\exp\left[ik\int_{o}^{z}\Delta_b\ (z')\,dz'\right] \end{aligned} \tag{10}$$

where $\Delta_a^{\pm}$, $\Delta_b^{\pm}$ are the elements of the diagonal matrix $\underline{\underline{\Delta}}_D$.
The amplitudes a^{-}, b^{-}, of the backword propagating waves are identically zero, since reflection is neglected.
For light normally incident on slab - type samples of a cholesteric liquid crystal, it is possible to get in this way the same results obtained by Mauguin [2] in the limit of

large enough pitches (Mauguin or adiabatic limit). In this case Δ_a and Δ_b are the extraordinary and ordinary refractive indexes n_e e n_o respectively; ψ_a^+ (ψ_b^+) represents a wave whose electric vector is everywhere parallel (perpendicular) to the optical axis, rigidly following the rotation of the director.

In this particular problem, considering as independent variables the amplitudes a^+, b^+, a^-, b^- of the characteristic vectors ψ_a^+ , ψ_b^+ , ψ_a^- , ψ_b^- is practically equivalent to analyze the electromagnetic wave in a new reference system which rigidly follows the rotation of the optical axis. For more general rotations of the dielectric tensor the use of eq. (8) offers similar advantages, and the GOA can be considered as an extension of the adiabatic limit to such cases.

3.2 Generalized geometrical optics approximation (GGOA).

For an isotropic medium with a continuous variation of the refractive index the GOA is valid if both diffraction and reflection have negligible effects. For anisotropic media, the GOA further implies that the waves ψ_a^+ and ψ_b^+ propagate independently. It is interesting to find out the validity limits of these approximations.

In most experiments with liquid crystals the cross-section dimensions of the sample and of the incident light beam are such that diffraction is indeed negligible. In order to evaluate the magnitude of the other two effects we consider the propagation of the a^+ component through a distance d such that the components of the tensor $\underline{\underline{\varepsilon}}$ are significantly

and monotonically changed, owing to a director rotation. At any layer dz this component is generally generating the other three components ψ_b^+, ψ_a^-, ψ_b^-.
The corresponding amplitudes a^+, a^-, b^- are however not monotonically increasing with increasing z, because the waves generated by different layers are out of phase. These amplitudes are actually negligible if the maximum phase difference, between the layers separated by a distance d, is large with respect to 2π. This gives for b^+ the condition:

$$\int_0^d \left| \Delta_a^+ - \Delta_b^+ \right| dz \gg \lambda \tag{11}$$

Similar conditions are found for the amplitudes a^-, b^- of the reflected waves, and are generally weaker than condition (11). As an example, let us consider the case of normal incidence of light on an uniaxial medium whose optical axis is nearly perpendicular to z. These conditions are approximatively given by

$$d \gg \lambda / (n_e - n_o) \tag{12}$$

for the ψ_b^+ wave, and (13)

$$d \gg \lambda / (n_e + n_o)$$

for the reflected waves ψ_a^+ and ψ_b^-.

For many experimental set-ups and devices (as e.g. the twisted and supertwisted nematic cells) the condition (13) is well satisfied, while the condition (12) is not. It is therefore interesting to find out an approximation which takes into account the coupling between the forward-propagating waves and neglects reflection. This aim is very easily achieved by making use of the formalism outlined above. In fact, the matrix $\underline{\underline{T}}^{-1}\ d\underline{\underline{T}}/dz$ is written as a sum of the two submatrices:

$$C_o = \begin{pmatrix} C_{11} & C_{12} & & \\ C_{12} & C_{22} & 0 & 0 \\ 0 & 0 & C_{33} & C_{34} \\ 0 & 0 & C_{43} & C_{44} \end{pmatrix}, \quad C_1 = \begin{pmatrix} 0 & 0 & C_{13} & C_{14} \\ 0 & 0 & C_{23} & C_{24} \\ C_{31} & C_{32} & 0 & 0 \\ C_{41} & C_{42} & 0 & 0 \end{pmatrix} \tag{14}$$

and the matrix $\underline{\underline{C}}_1$ (whose elements give reflection) is simply dropped, whereas Co is retained in the propagation equations (8). The set of equations (8) splits now into two independent subsets. Since reflection is neglected, only the first set is of interest. This may be written:

$$\frac{d}{dz}\begin{pmatrix} a^+ \\ b^+ \end{pmatrix} = \left[ik \begin{pmatrix} \Delta a^+ & 0 \\ 0 & \Delta\, {}^+_b \end{pmatrix} - \begin{pmatrix} C_{11} & C_{12} \\ C_{21} & C_{22} \end{pmatrix} \right] \begin{pmatrix} a^+ \\ b^+ \end{pmatrix} \tag{15}$$

This approximation is a possible extension of the GOA for isotropic media, in the sense that both of them neglect diffraction and reflection. In the following, it will be referred to as GGOA (generalized geometric optics approximation); we note that the same expression is used in ref. 20 with a different meaning). The GGOA is particularly suitable for the description of the polarization state of the transmitted wave, since the coupling between the forward-propagating waves if fully taken into account, and reflection can change this state only through second order effects, as obvious.

For normally incident light the information content of the parameters a^+, b^+ and of the Jones vector components is the same. The column vector with components (a^+,b^+) can therefore be considered as a generalization of the Jones vector. This allows one to discuss the results given in the above quoted references in the framework of the formalism developed here.

4 Comments and discussion.

The most important results obtained for aperiodic structures in the references quoted in section 1 can now be discussed in the terms of the formalism developed in sections 2 and 3. Most papers develop approximations which are essentially equivalent to GGOA, and only consider the case of normally incident light [11,12,14,16].

The case of oblique incidence, which generally requires the use of quite heavy expressions, has been considered only for the particular case of a locally uniaxial film with the optical axis everywhere orthogonal to the surface normal [15].

The GOA approximation has been applied to the following two particular cases:

1) The medium is locally uniaxial, and the optical axis always lies in the incidence plane [20].

The extraordinary and ordinary waves are fully decoupled, and only the first one requires approximate expressions, as obvious.

2) The medium is locally uniaxial and only the case of normal incidence is considered, by means of the use of the Stokes vector formalism [13].

All the above results, are implicitly contained in equations 5, 16 and 15. In order to obtain a formal expression of these results, both the characteristic values Δ_a^+, Δ_b^+, Δ_a^-, Δ_b^- and the corresponding characteristic vectors must be given as functions of the angles defining the dielectric tensor $\underline{\underline{\varepsilon}}(z)$. They are given in the Appendix for the particular case of uniaxial media.

<u>Appendix</u> - Characteristic values and vectors of the propagation matrix for uniaxial crystals.

The matrix for a nonmagnetic uniaxial medium without losses and optical activity is [19]

$$\underline{\underline{\Delta}} = \begin{pmatrix} Mc & 1-m^2/\varepsilon_{33} & Ms & 0 \\ n_o^2 s^2 + n^2 c^2 & Mc & (n^2-n_o^2)sc & 0 \\ 0 & 0 & 0 & 1 \\ (n^2-n_o^2)sc & Ms & n_o^2c^2+n^2s^2-m^2 & 0 \end{pmatrix} \quad \text{A1}$$

where:

$c = \cos\varphi \quad , \quad s = \sin\varphi$

$m = k_x/k$, $M = m\cos\vartheta \, \sin\vartheta \, (n_o^2-n_e^2)/\varepsilon_{33}$

$$\varepsilon_{33} = n_e^2 \cos^2\vartheta + n_o^2 \sin^2\vartheta \, , \, n^2 = n_o^2 n_e^2/\varepsilon_{33} \quad \text{A2}$$

$$\Delta_o = (n_o^2-m^2)^{\frac{1}{2}}; \Delta_e = \left[-\frac{m^2}{\varepsilon_{33}} (n_o^2 \sin^2\varphi + n^2\cos^2\varphi) + n^2 \right]^{\frac{1}{2}}$$

where ϑ, φ are the polar angles defining the direction of the optical axis, n_e and n_o are the extraordinary and ordinary refractive indexes, respectively, and the use of Δ_o, Δ_e will be apparent below.
We recall that z is the polar axis and (x,z) the incidence plane.

The characteristic equation for $\underline{\underline{\Delta}}$,

$$\det(\underline{\underline{\Delta}} - \Delta \underline{\underline{1}}) = 0 \quad \text{A3}$$

is an equation of fourth degree for the characteristic values Δ of $\underline{\underline{\Delta}}$. For uniaxial media the two roots of this equation, corresponding to forward - and backward - propagating ordinary waves, are opposite and well known. By taking into account this fact, it is easily shown that eq. A3 can be cast in the form:

$$\left[\Delta^2-n_o^2+m^2\right]\left[\Delta^2-2M\cos\varphi\Delta+M^2\cos^2\varphi-\Delta_e^2\right]=0 \qquad \text{A4}$$

This gives:

$$\Delta_a^{+,-}=M\cos\varphi\pm\Delta_e \quad , \quad \Delta_b^{+,-}=\pm\Delta_o \qquad \text{A5}$$

The corresponding characteristic vectors are easily found to be:

$$\psi_a^{\pm}=N_a^{\pm}\begin{pmatrix}(n^2-n_o^2)(1-m^2/\varepsilon_{33})\cos\varphi\pm M\\ M(n_o^2\sin^2\varphi+n^2\cos^2\varphi)\pm(n^2-n_o^2)\Delta_e\cos\varphi\\ (n^2-n_o^2)\sin\varphi\\ (n^2-n_o^2)M\sin\varphi\cos\varphi\pm(n^2-n_o^2)\Delta_e\sin\varphi\end{pmatrix}$$

A6

$$\psi_b^{\pm}=N_b^{\pm}\begin{pmatrix}\pm(n_o^2-n^2)\Delta_o\sin\varphi\\ (n_o^2-n^2)n_o^2\sin\varphi\\ n_o^2M\pm(n^2-n_o^2)\Delta_o\cos\varphi\\ (n^2-n_o^2)\Delta_o^2\cos\varphi\pm Mn_o^2\Delta_o\end{pmatrix}$$

Where N_a^+, N_b^+, N_a^-, N_b are normalization constants. A suitable choice for these parameters is the one which gives

unit energy flux for unit area of the film, i.e. unit z-component of the Poynting vector.
We recall that ψ_a^+, ψ_b^+, ψ_a^-, ψ_b^- are the characteristic vectors for a homogeneous medium and that they define the trasformation matrix $\underline{\underline{T}}$, whose columns are proportional to the column vectors which define ψ_a^+, ψ_b^+, ψ_a^-, ψ_b^-, respectively.

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