

The cholesteric liquid crystal as a spatially dispersive medium

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MS received 6 November 1973

Abstract. A medium with periodic variation of the dielectric tensor is considered. It is assumed to have helical symmetry. The response to external fields is described by the most general linear law—that is, by the methods of spatial dispersion theory. The propagation of a wave is described by a mutually consistent electric field and polarisation. It is shown that the presence of the medium produces changes in the polarisation and wave vector of the electric field, and the selection rules governing these changes are derived from symmetry. The results generalise previous work on the Oseen model for a cholesteric liquid crystal to the case when the molecules are not perpendicular to the helical axis. This can arise in an external magnetic field applied along this axis.

Keywords. Liquid crystal; dispersion theory.

Introduction

This paper considers the optical properties of a medium with helical symmetry. An important practical example of such a medium is the cholesteric liquid crystal, which has received much theoretical and experimental attention. Discrete and continuum models have been proposed and discussed in the literature, which is cited in section 1. They have been shown to explain experimental results. Here, the problem is investigated using spatial dispersion theory, which is outlined in section 2. Only the helical symmetry of the medium and a linear response to the external field are assumed.

The main results of the paper are that the helical symmetry severely restricts the transformations of wave vector and polarisation which can take place. This enables the propagation of waves along the axis to be described in a general form—*viz.*, as a superposition of left and right circular waves whose wave vectors are related by the Bragg condition, along with a wave polarised along the axis. Earlier work, which solved the problem with two circular waves, is thus generalised. The additional longitudinal wave will appear in a cholesteric liquid crystal placed in a strong axial magnetic field.

Section 1 briefly introduces a continuum model with which the final results can be compared—the generalised Oseen model. Section 3 presents the derivation of selection rules governing the changes in wave-vector and polarisation which the fields undergo on interacting with the medium. It also explains how these rules determine the nature of the normal waves in the medium, and shows how they are related to the Oseen model of section 1.

1. The generalised Oseen model (Oseen 1933)

The molecules in any small region of the cholesteric liquid crystal are oriented preferentially around a direction which is usually described by a vector called the director. The director is constant on planes perpendicular to the helical axis (say z) and rotates around this axis as we move along it. In the absence of external fields it is normal to the z -axis, while a sufficiently strong magnetic field along z is predicted to bring it into an inclined configuration (Leslie 1970).

We take the dielectric tensor at the plane $z = 0$ to have general principal axes so that it has the form

$$\epsilon(z=0) \equiv \begin{bmatrix} \epsilon_{11} & \epsilon_{12} & \epsilon_{13} \\ \epsilon_{12} & \epsilon_{22} & \epsilon_{23} \\ \epsilon_{13} & \epsilon_{23} & \epsilon_{33} \end{bmatrix} \quad (1)$$

Because of the helical symmetry, we obtain $\hat{\epsilon}(z)$ for a general value of z by using the matrix describing a rotation by the angle $-qz$ in the xy plane (figure 1) to transform eq. 1. Here q is the angle of rotation per unit distance along the z -axis. Defining the pitch P as the distance needed for a rotation of 2π to take place, we get $qP = 2\pi$ or $q = 2\pi/P$. The result of the transformation is given below. All tensor components are functions of z .

$$\begin{aligned} \epsilon_{zz} &= \bar{\epsilon} + \delta \cos 2qz - \epsilon_{12} \sin 2qz \\ \epsilon_{xy} &= \epsilon_{yx} = \delta \sin 2qz + \epsilon_{12} \cos 2qz \\ \epsilon_{zx} &= \epsilon_{xz} = \epsilon_{13} \cos qz - \epsilon_{23} \sin qz \\ \epsilon_{yy} &= \bar{\epsilon} - \delta \cos 2qz + \epsilon_{12} \sin 2qz \\ \epsilon_{yz} &= \epsilon_{zy} = \epsilon_{13} \sin qz + \epsilon_{23} \cos qz \\ \epsilon_{xx} &= \epsilon_{33} \end{aligned} \quad (2)$$

We have defined

$$\bar{\epsilon} = \frac{\epsilon_{11} + \epsilon_{22}}{2}, \quad \delta = \frac{\epsilon_{11} - \epsilon_{22}}{2}$$

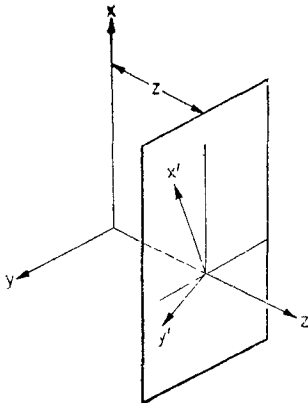


Figure 1. Helical symmetry. The properties of the medium are identical whether viewed from the origin in the xyz system of axes, or from a point displaced by z along the z -axis, using $x'y'z'$ as axes of reference. $x'y'$ are tilted with respect to xy by an angle qz . To transform back to xy we must rotate by $-qz$.

The Oseen model (OM) in its usual form omits the ϵ_{13} and ϵ_{23} terms by choosing one of the principal axes along z in eq. 1. This describes molecules oriented perpendicular to z . The ϵ_{12} term can be eliminated in any case by a suitable choice of the origin for z .

Equation 2 enables us to anticipate a property of the general solution which is obtained in section 4. The terms in $\sin 2qz$ and $\cos 2qz$ modulate the electric field, introducing wave vectors differing from the original by $\pm 2q$. Similarly $\cos qz$ and $\sin qz$ cause changes by $\pm q$. These latter terms link the x and y polarisation to the z polarisation.

The OM in the restricted form was exactly solved for waves propagating along the axis by Kats (1970), Marathay (1971) and the author (Nityananda 1972; *see also* Nityananda and Ramaseshan 1971). The last paper also shows that the discrete model (pile of birefringent plates) treated by Chandrasekhar and Srinivasa Rao (1968)* is equivalent to the OM.

In this paper, the assumption of a local dielectric tensor which is a periodic function of position is removed. The medium is characterised solely by its helical symmetry and a linear response to an applied electromagnetic field. This suffices to explain the optical properties and, surprisingly, the exact solution of the normal incidence problem is still possible, even when terms equivalent to the ϵ_{13} and ϵ_{23} of eq. 2 are retained. This solution describes wave propagation in a cholesteric liquid crystal with the long axes of the molecules tilted towards the axis of the helix, rather than normal to it. Such a conical ordering may be produced by a static magnetic field applied along the axis (Leslie 1970).

This configuration is energetically favourable provided the diamagnetic susceptibility is numerically greater perpendicular to molecular axis, so that this direction in turn tends to be perpendicular to the field. This is not so in cholesteryl chloride and cholesteryl myristate, two of the commonly studied cholesteric materials. Therefore, a special choice of material seems necessary before such conical ordering can be achieved.†

2. Methods of spatial dispersion theory

Since the methods of spatial dispersion theory have not been applied in this field they are summarised below. Details and references can be found in the review by Ginzburg and Agranovich (1966).

Dispersion usually connotes the frequency dependence of the dielectric response, ion, equivalently, the failure of the induced polarisation to follow the variation of the applied field in *time*. For example, if we apply an impulsive field

$$E(t) = \delta(t - t_0) \quad (3)$$

* For improvements and corrections, *see* Chandrasekhar and Shashidhara Prasad (1971); Chandrasekhar and Ranganath (1973).

† I thank the referee for this remark.

the polarisation is not itself impulsive but grows and decays according to a definite law

$$P(t) = K(t - t_0) \quad (4)$$

Figure 2 shows $K(t - t_0)$ for a damped oscillator. The response to an impulsive field determines, in principle, the response to an arbitrary field $E(t)$ since this can be regarded as a continuous superposition of impulses

$$E(t) = \int_{-\infty}^{+\infty} E(t') \delta(t - t') dt' \quad (5)$$

Using the linear property of the response, $P(t)$ is obtained by replacing $\delta(t - t')$ in eq. 5 by $K(t - t')$ which is zero for $t < t'$. We get

$$P(t) = \int_{-\infty}^t K(t - t') E(t') dt' \quad (6)$$

We can use eq. 4 to determine the response to a harmonically varying field

$$E(t') = E_0 e^{-i\omega t'}$$

We get

$$\begin{aligned} P(t) &= \int_{-\infty}^t K(t - t') E_0 e^{-i\omega t'} dt' \\ &= - \int_{\infty}^0 K(\tau) E_0 e^{-i\omega(t-\tau)} d\tau \text{ putting } t' = t - \tau \\ &= \alpha(\omega) E_0 e^{-i\omega t} \end{aligned} \quad (7)$$

where

$$\alpha(\omega) = \int_0^{\infty} K(\tau) e^{i\omega\tau} d\tau$$

The response is linear in E_0 with a frequency dependent coefficient $\alpha(\omega)$ related to K . Equations (3) to (7), represent a well known approach to frequency dependence of the polarisability (Landau and Lifshitz 1968) which has been repeated here for comparison with spatial dispersion which is described below.

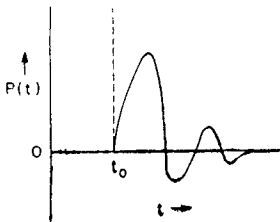


Figure 2. The response of a damped oscillator to an impulsive electric field applied at $t = t_0$

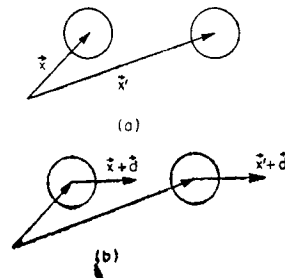


Figure 3. (a) A configuration of x and x' in which the response function $K(x, x')$ is appreciable, (b) A configuration with the same value of $x - x'$ in which $K(x, x')$ is much smaller.

Proceeding similarly, a localised electric field

$$E(\mathbf{x}) = E_0 \delta(\mathbf{x} - \mathbf{x}_0) \quad (8a)$$

will produce a polarisation given in general by eq. 8 b and not a localised polarisation

$$P(\mathbf{x}') = K(\mathbf{x}', \mathbf{x}_0) E(\mathbf{x}_0) \quad (8b)$$

We have explicitly brought out the vector nature of E and P and the tensor nature of K in this section. Further, K depends on \mathbf{x}' and \mathbf{x}_0 individually and not solely on their difference, in contrast to the case of temporal dispersion. That is because we are not assuming the homogeneity of space, while eq. (4) assumes the homogeneity of time. An example clarifying this is given in figure 3. Figure 3 a shows the application of a localised field at \mathbf{x} where an atom is located. The atom is polarised and produces a local field at its neighbour which is at \mathbf{x}' . This gives rise to some value of $K(\mathbf{x}', \mathbf{x})$. However, if we translate both \mathbf{x} and \mathbf{x}' by d , as shown in figure 3 b, the whole effect is very much smaller because polarisable matter is absent at \mathbf{x} and \mathbf{x}' , even though the difference $\mathbf{x} - \mathbf{x}'$ is the same as before. This example can be used to make the point that there are two situations which can be treated by the formalism of spatial dispersion theory—inhomogeneity and local field effects. The generalised Oseen model mentioned in the introduction clearly introduces inhomogeneity effects, and it corresponds to taking

$$K(\mathbf{x}, \mathbf{x}') = F(\mathbf{x}) \delta(\mathbf{x} - \mathbf{x}')$$

It does not explicitly include local field effects. We expect inhomogeneity effects to be appreciable if the applied electric field has significant variation over a length characterising the spatial variation of the system. This could be the interatomic spacing ' a ' in the case of crystals, and in crystal optics a/λ can be taken as the small parameter characterising spatial dispersion effects. Optical activity is of the first order in this parameter, and the birefringence of cubic crystals of the second order (Agranovich and Ginzburg 1965, Pastrnak and Vedam 1971). In the cholesteric liquid crystal the pitch P determines the inhomogeneity and the 'small' parameter P/λ is of the order of unity. This is the motivation for considering it a spatially dispersive medium.

It is convenient, as in the case of temporal dispersion, to consider the response to a field $E(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{x}}$ which has a periodic variation in space. We obtain

$$\begin{aligned} P(\mathbf{x}) &= \int K(\mathbf{x}, \mathbf{x}') E(\mathbf{x}') d\mathbf{x}' \\ &= \int K(\mathbf{x}, \mathbf{x}') E(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{x}'} d\mathbf{x}' \end{aligned} \quad (9)$$

We Fourier analyse $P(\mathbf{x})$ to obtain $P(\mathbf{k}')$

$$\begin{aligned} P(\mathbf{k}') &= \int P(\mathbf{x}) e^{-i\mathbf{k}' \cdot \mathbf{x}} d\mathbf{x} \\ &= \iint K(\mathbf{x}, \mathbf{x}') E(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{x}'} e^{-i\mathbf{k}' \cdot \mathbf{x}} d\mathbf{x}' d\mathbf{x} \\ &= L(\mathbf{k}', \mathbf{k}) E(\mathbf{k}) \end{aligned} \quad (10)$$

where

$$L(\mathbf{k}', \mathbf{k}) = \iint K(\mathbf{x}, \mathbf{x}') e^{i\mathbf{k} \cdot \mathbf{x}'} e^{-i\mathbf{k}' \cdot \mathbf{x}} d\mathbf{x} d\mathbf{x}' \quad (11)$$

$P(\mathbf{k}')$ measures the polarisation of wave vector \mathbf{k}' generated by an applied field of wave vector \mathbf{k} . The reason for the appearance of a new wave vector in the induced polarisation is that the medium is not homogeneous and modulates the applied field $E(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{x}}$ with its own periodicities. For a homogeneous medium, $K(\mathbf{x}, \mathbf{x}')$ is a function of the difference $\mathbf{x} - \mathbf{x}'$ and in this case $L(\mathbf{k}', \mathbf{k})$ is seen to vanish for $\mathbf{k}' \neq \mathbf{k}$. This is the usual assumption in crystal optics (Ginzburg and Agranovich 1966), but the ability of the liquid crystal to change the wave vector of an incident wave is central to its optical properties, so we retain $L(\mathbf{k}, \mathbf{k}')$ in full generality.

We now consider the role of $L(\mathbf{k}, \mathbf{k}')$ in determining the nature of the wave propagated in the medium. If we seek a normal wave—one with a harmonic time dependence $\exp(-i\omega t)$ —we can consider the wave equation in the form

$$\nabla \times (\nabla \times E) = \frac{\omega^2}{c^2} (E + 4\pi P) \quad (12)$$

This can be regarded as determining the electric field E radiated by a polarisation P . To this we add the constitutive equation in the form (9) or (10). The determination of a normal wave is thus seen to be a self-consistency problem—the field radiated by the polarisation P must be precisely enough to maintain it (Ewald 1966). If we choose to express E and P in terms of Fourier components as in (10) and introduce the result into (12), the left and right sides become homogeneous linear expressions in the components of $E(\mathbf{k})$ for various \mathbf{k} 's. To determine $E(\mathbf{k})$ we thus have to solve a secular equation which gives the dispersion relation for the normal waves, as well as the values of $E(\mathbf{k})$ for various \mathbf{k} 's. An example of this procedure can be found in the papers on the Oseen model by Kats (1970) and by the author (Nityananda 1973); in these treatments the normal wave was a superposition of left and right circular waves with different wave vectors.

3. Selection rules for $L(\mathbf{k}, \mathbf{k}')$ in the presence of helical symmetry

We first need a symmetry property of K , viz.

$$K_{ij}(\mathbf{x}, \mathbf{x}') = K_{ji}(\mathbf{x}', \mathbf{x}) \quad (13)$$

The physical meaning of this relation is illustrated in figure 4. In figure 4 *a*, an electric field is applied at \mathbf{x}' along the cartesian axis j and the response P measured at \mathbf{x} along the cartesian axis i . This represents $K_{ij}(\mathbf{x}, \mathbf{x}')$. Figure 4 *b* similarly represents $K_{ji}(\mathbf{x}', \mathbf{x})$ and eq. (13) asserts that the responses in these two cases are numerically equal. Equation (13) is only a particular case of equation (14) which includes temporal dispersion as well

$$K_{ij}(\mathbf{x}, t; \mathbf{x}', t') = K_{ji}(\mathbf{x}', t; \mathbf{x}, t') \quad (14)$$

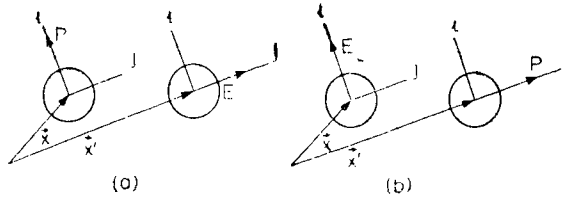


Figure 4. (a) The configuration defining $K_{ij}(x, x')$, (b) The configuration defining $K_{ji}(x', x)$. In both cases the vector P indicates that component which is being measured, not the entire polarisation generated.

Equation (14) can be explicitly derived from the Kubo formula (Kubo 1957) expressing response functions in terms of correlation functions, but can also be regarded as a descendant of the Onsager reciprocity relations, or, in the case where there is no dissipation, even as a consequence of the interchangeability of source and field points in electrostatic problems. Equation (13) can be combined with eq. (11) to give

$$L_{ij}(k, k') = L_{ji}(-k', -k) = L_{ji}^*(k', k) \quad (15)$$

the last equality assuming that K is real, *i.e.*, temporal dispersion effects are neglected.

Now we can exploit the helical symmetry of the medium. It is left invariant under (i) arbitrary translations by vectors $\lambda\hat{i} + \mu\hat{j}$ in the xy plane ($\hat{i}\hat{j}\hat{k}$ are the usual unit vectors), (ii) Translation by $t\hat{k}$ along z followed by rotation by an angle $\theta = (t/P)2\pi$ about the z -axis. A particular case is simply translation by $nP\hat{k}$ where n is any integer, without rotation.

(i) Replacing x and x' by $x + \lambda\hat{i} + \mu\hat{j}$ and $x' + \lambda\hat{i} + \mu\hat{j}$ should not alter the value of K . This implies

$$\begin{aligned} L_{ij}(k, k') &= \int K_{ij}(x, x') e^{-ik \cdot x} e^{ik' \cdot x'} dx dx' \text{ from (11)} \\ &= \int K_{ij}(x + \lambda\hat{i} + \mu\hat{j}, x' + \lambda\hat{i} + \mu\hat{j}) e^{-ik \cdot x} e^{ik' \cdot x'} dx dx' \\ &= e^{i(k-k') \cdot (\lambda\hat{i} + \mu\hat{j})} \int \int K_{ij}(u, v) e^{i(k' \cdot v - k \cdot u)} du dv \\ &= e^{i(k-k') \cdot (\lambda\hat{i} + \mu\hat{j})} L_{ij}(k, k') \end{aligned}$$

The exponential can be equal to unity for all values of λ and μ only if $(k - k') \cdot \hat{i} = (k - k') \cdot \hat{j} = 0$, that is, $k - k'$ has no component in the xy plane. This selection rule is clearly analogous to the conservation of the x and y components of momentum and is a consequence of the homogeneity in the xy plane.

(ii) We first consider translations by $nP\hat{k}$ which will give us, as the condition

$$e^{i(k-k') \cdot nP\hat{k}} = 1$$

for all integral n . Hence

$$(k - k') \cdot \hat{k} = 2m\pi/P$$

Combining this with the condition derived in (i) and introducing the reciprocal lattice vector

$$G = \frac{2\pi}{P} \hat{k}$$

we get

$$k - k' = mG.$$

Now we consider the operation of rotation by θ about the z axis. The components of E and P are transformed to new values

$$\begin{aligned} \bar{E}_i &= R_{ij} E_j \\ R &\equiv \begin{bmatrix} \cos \theta & \sin \theta & 0 \\ -\sin \theta & \cos \theta & 0 \\ 0 & 0 & 1 \end{bmatrix} \end{aligned} \quad (16)$$

The new tensor \bar{K} relating E and P is given by

$$\bar{K} = RKR^{-1} \quad \text{or} \quad K_{mn} = R_{mi} K_{ij} R_{jn}^{-1}$$

L also transforms similarly, since the double Fourier transformation (11) relating L and K does not disturb the tensor indices i and j . Under a translation of $(\theta/2\pi)P$ and a rotation by θ the medium must remain invariant. Under the translation we get

$$L' = \exp \{-i(k-k') \cdot \hat{k} \theta P/2\pi\} L = e^{-in\theta} L$$

using

$$k - k' = nG, \quad G \cdot \hat{k} = 2\pi/P$$

Now if we rotate by θ about z we should recover the original L .

$$L = e^{-in\theta} R L R^{-1}$$

Setting $\sin \theta = s$, $\cos \theta = c$, we get

$$L_{11} = e^{-in\theta} (c^2 L_{11} + s^2 L_{22} + cs(L_{12} + L_{21})), \text{ etc.} \quad (17)$$

Equation (17) has to be fulfilled as an identity in θ . Since the left side is independent of θ , only those terms on the right which are also independent of θ can be non-vanishing. Since the power of $e^{i\theta}$ in the matrix on the RHS of eq. (17) lies between -2 and $+2$, it is only for n in the same range that we can get a term independent of θ on the RHS. Hence, in addition to the selection rule $k - k' = nG$ which follows from periodicity, we obtain further the result $n = 0, \pm 1, \pm 2$ on account

of the continuous helical symmetry. For each of the values of n , the non-vanishing coefficients and the relations between them can be found from eq. (17). Only one illustration of this routine procedure is given, and the results are collected later.

(i) $n = \pm 2$: Equating the 11 component on both sides in eq. (17)

$$\begin{aligned} L_{11} (\cos 2\theta \pm i \sin 2\theta) &= L_{11} \cos^2 \theta + L_{22} \sin^2 \theta + (L_{12} + L_{21}) \sin \theta \cos \theta \\ &= \left(\frac{L_{11} + L_{22}}{2} \right) + \left(\frac{L_{11} - L_{22}}{2} \right) \cos 2\theta + \left(\frac{L_{12} + L_{21}}{2} \right) \sin 2\theta \\ \therefore L_{11} &= \frac{L_{11} - L_{22}}{2}, \quad L_{22} = -L_{11}, \quad \frac{L_{12} + L_{21}}{2} = \pm iL_{11} \end{aligned}$$

Similarly we can compare the 12, 13... etc., components. The results concerning the nonvanishing components are:

$$\underline{n = \pm 2}: \quad L_{22} = -L_{11}, \quad L_{12} = L_{21} = \pm iL_{11}$$

$$\underline{n = \pm 1}: \quad L_{32} = \pm iL_{21}, \quad L_{23} = \pm iL_{13}$$

$$\underline{n = 0}: \quad L_{11} = L_{22}, \quad L_{33} \text{ nonvanishing.}$$

The remaining components vanish.

The value of n will be used as a superscript if necessary. We introduce a notation for $L^{(n)}(k, k')$ which treats k and k' more symmetrically by writing

$$k = q + \frac{nG}{2}; \quad k' = q - \frac{nG}{2}$$

L is now a function of the average wave vector q and the index n . The symmetry condition eq. (15) now reads

$$L_{ij}^{(n)}(q) = L_{ji}^{*(-n)}(q)$$

We now introduce new variables

$$E_{\pm} = \frac{E_x \pm iE_y}{\sqrt{2}}$$

in place of the x and y components of the electric field. These describe two opposite circular polarisations. We can transform the components of the tensor L so that it can operate on the new variables.

Since

$$\begin{bmatrix} E_+ \\ E_- \\ E_z \end{bmatrix} = \begin{bmatrix} 1/\sqrt{2} & i/\sqrt{2} & 0 \\ 1/\sqrt{2} & -i/\sqrt{2} & 0 \\ 0 & 0 & 1 \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} = [A] \begin{bmatrix} E_+ \\ E_- \\ E_z \end{bmatrix}$$

is the transformation law for the field components, the transformation law for L is $\tilde{L} = ALA^{-1}$. The tilde denotes a quantity expressed in the circular wave representation, *i.e.*, operating on the \pm and z components. The transformed L 's are given below.

$n = 0$

$$\tilde{\mathbf{L}}^0 \equiv \begin{bmatrix} \tilde{L}_{11} & 0 & 0 \\ 0 & \tilde{L}_{22} & 0 \\ 0 & 0 & \tilde{L}_{33} \end{bmatrix}$$

 $n = \pm 1$

$$\tilde{\mathbf{L}}^{(+1)} \equiv \begin{bmatrix} 0 & 0 & \tilde{L}_{13} \\ 0 & 0 & 0 \\ 0 & \tilde{L}_{31} & 0 \end{bmatrix}$$

$$\tilde{\mathbf{L}}^{(-1)} \equiv \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \tilde{L}_{31}^* \\ \tilde{L}_{13}^* & 0 & 0 \end{bmatrix}$$

 $n = \pm 2$

$$\tilde{\mathbf{L}}^{(+2)} \equiv \begin{bmatrix} 0 & 2L_{11} & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

$$\tilde{\mathbf{L}}^{(-2)} \equiv \begin{bmatrix} 0 & 0 & 0 \\ 2L_{11}^* & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

We can physically interpret these relations as follows: The $n = +2$ component tells us that if we impress an electric field E_- , i.e., a left circular wave propagating along $+z$, then a polarisation, P_+ will be generated with wave vector increased by $2G$. Similarly an applied wave E_+ will generate a polarisation P_- with wave vector reduced by $2G$. This is in agreement with the results found for the restricted Oseen model (Kats 1970; Rajaram Nityananda 1972). This property made it possible to find a normal wave made up of left and right circular waves differing by $2G$ in wave vector. Now, however, we have the $n = \pm 1$ terms which can convert $+$ and $-$ polarisations into 'z' polarisation with a change of wave vector by G .

The allowed changes are represented in a graphical scheme in figure 5. There are no other possibilities of generating new wave vectors or polarisations. Thus for this case, a normal wave can be built up of a left circular wave and a right circular wave accompanied by a longitudinal wave (described by the E_z component). The dispersion relation and the coefficients of the three components must be determined from a 3×3 secular equation by methods quite similar to those used for the Oseen model. The fact that these three waves form a closed set enables us to build up a normal wave from them and solve a third order secular equation.* This property fails when we go to oblique incidence. The operator $\nabla \times (\nabla \times)$ on the left side of the wave equation (12) can generate $+$ and $-$ components from E_z with the same wave vector, and this immediately takes us out of the scheme illustrated earlier.

* To avoid misunderstanding, we note that the equation is still quartic in q and gives us 4 normal waves, two in each direction, as before.

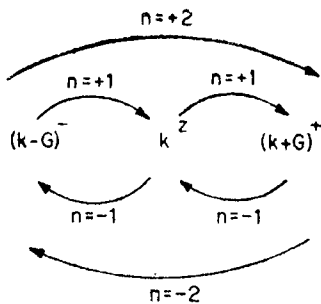


Figure 5. A graphical representation of the selection rules for wave vector and polarisation. The latter is denoted by +, -, and z written as a superscript to the wave vector. There are no other allowed transitions; so a suitable combination of these three can be chosen as a normal wave.

4. Conclusions

The constitutive relation for a medium with helical symmetry has been constructed using only the linear response and symmetry properties. The various nonvanishing coefficients have been found in the most general case, and their effect on the nature of the normal waves which can propagate in this medium has been discussed. Earlier results on the Oseen model for a cholesteric liquid crystal emerge as special cases. Even when the principal axes of the dielectric ellipsoid have no special orientation which respect to the helical axes, the normal waves can be constructed from two circular waves and a longitudinal wave. This solution may be of interest in describing the optical properties of a cholesteric liquid crystal placed in a strong magnetic field parallel to its axis, subject to the qualifications made in section 1.

Note added in proof

Recently D W Berreman (1973 *Mol. Cryst. Liquid Cryst.* **22** 175) has used the Oseen model with an arbitrary orientation of the principal axes (as in section 1) to calculate the optical properties of the twisted smectic C phase. His approach to the problem is numerical. The results of the present paper should be applicable to this problem as well.

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