Dispersion relation for propagation of light in cholesteric liquid crystals

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A general theory of the light propagation in periodic structures characterized by a uniform rotation of the dielectric tensor about a given axis is presented. Starting from a fundamental approach of Dreher and Meier, which is mostly numerical, an analytical solution of the characteristic equation has been found which can be used to calculate the wave vectors as a function of ω and of the incidence angle θ_i. The electromagnetic wave is described as a superposition of elementary modes having the form of Bloch waves. Each elementary mode is represented by a sum of plane waves elliptically polarized, whose wave vectors are the roots of the characteristic equation. The analysis of the solutions of such an equation allows us to draw a more complete map of the stability and instability regions for light propagation in helical structures than the ones currently available in the literature. The coexistence of two distinct modes, with different polarization states, determines the shape of the stability map. Each mode presents a series of Bragg instabilities. Between the two Bragg instabilities of the same order a further instability exists which is common to both modes and does not satisfy the Bragg conditions. All instability bands, with the exception of only one of the first order, vanish at normal incidence. This occurs for any value of the optical anisotropy and is a peculiarity of perfectly ordered helical structures. The bandwidth increases with θ_i, and overlapping may occur. Typical plots of dispersion curves and attenuation constants are reported. Finally, we compute the intensity and the polarization state of the light reflected from a thin film, in order to clarify the controversial point about the structure—doublet or triplet—of the higher-order reflection bands.

I. INTRODUCTION

The discovery of cholesteric liquid crystals stimulated the study of the propagation of electromagnetic waves in helicoidal structures having a definite periodicity. Exact analytical solutions to this problem have been found only for the particular case of a plane wave propagating in the direction of the helix axis.[1,2]

So far no simple analytic solutions have been presented in the literature for the case of oblique incidence. In such a case the different parameters of physical interest are generally found by using a numerical approach, such as, for instance, the method of the propagation matrices in stratified media.[3] In the present paper a different type of approach is utilized, similar to the one used by Taupin[4] and by Dreher et al.[5] Such an approach gives very precise numerical results and, furthermore, allows a deeper

insight in the understanding of the light propagation in helicoidal structures. The method consists of finding out those solutions of the Maxwell equations which are in agreement with the Bloch-Floquet theorem. These solutions are the simplest propagating waves in the structure and will be referred to, in the following, as elementary modes of the propagating wave.

It is convenient to consider a semi-infinite sample on which a plane wave of a given angular frequency ω impinges with incidence angle θ_i. In analogy to what happens in a homogeneous anisotropic crystal, the incident wave splits within the sample into two separate elementary modes having different polarization states. The polarization states are now in general elliptic, and the shape of the ellipse changes along the helix axis. Actually, each mode can be represented as a superposition of plane waves having elliptic polarization and wave vectors $\vec{K}_n = \vec{K} + 2n\vec{q}$,

where n is an integer and $\vec{q}$ is a vector parallel to the helix axis whose modulus is related to the helix pitch p by the relation $q = 2\pi/p$. In the theory, amplitudes as well as polarization states of the plane-wave components will appear as unknowns in a set of algebraic homogeneous linear equations. The two characteristic values of $\vec{K}$ corresponding to the elementary modes of propagation can then be found by solving the determinantal equation of the set.

Taupin and Dreher et al. have solved numerically such a set of equations and found the conditions whereby the structure acts as a transmitting or a reflecting medium. In the present paper an analytical approach is adopted to the study of the determinantal equation. Starting from a set of algebraic equations similar to the one appearing in Dreher's paper it is shown that the determinant of the set can be expressed as a simple analytic function of $\vec{K}$. It is thus possible to obtain more complete results and to show that the results of Dreher et al. are not completely correct.

The developed method has been applied to calculate reflectance spectra from a thin film and shows a good agreement with the numerical calculations carried out by Berreman and Sheffer[6] with the 4×4 matrix technique.

II. PROPAGATION EQUATIONS

The medium is considered as a perfectly ordered cholesteric liquid crystal, whose macroscopic optical properties are described by a dielectric tensor uniformly spiraling around one of its principal axes. The latter is defined by the equation

$$\epsilon = \begin{bmatrix} \epsilon_m(1+\delta\cos2qz) & \epsilon_m\delta\sin2qz & 0 \\ \epsilon_m\delta\sin2qz & \epsilon_m(1-\delta\cos2qz) & 0 \\ 0 & 0 & \epsilon_3 \end{bmatrix} ,$$

(2.1)

where $\epsilon_m = (\epsilon_1 + \epsilon_2)/2$, $\delta = (\epsilon_1 - \epsilon_2)/(\epsilon_1 + \epsilon_2)$, and $\epsilon_1, \epsilon_2, \epsilon_3$ are the three principal values (a list of the principal symbols used in this paper is given at the end of this section).

According to Eq. (2.1) the structure is periodic along the z axis with a period $p/2 = \pi/q$ and has a complete translational symmetry along any direction perpendicular to the z axis. Owing to such a symmetry, Maxwell equations have solutions of the type

$$\begin{bmatrix} \vec{E}(\vec{r},t) \\ \vec{H}(\vec{r},t) \end{bmatrix} = \begin{bmatrix} \vec{E}(z) \\ \vec{H}(z) \end{bmatrix} e^{i(K_\perp x - \omega t)} .$$

(2.2)

Assuming that a plane wave of wave vector $\vec{K}_i$ penetrates within the cholesteric liquid crystal through a plane surface perpendicular to the z axis, the following relation between the incidence angle θ_i and $K_\perp$ can be written,

$$\sin\theta_i = \frac{K_\perp}{|\vec{K}_i|} = \frac{m}{n_i} ,$$

(2.3)

where $m = K_\perp c/\omega$, and n_i is the refraction index in the external medium.

Within the cholesteric liquid crystal a wave of the type (2.2) is obtained when the incidence plane of the wave is the plane $\{x,z\}$.

Upon substituting (2.2) in Maxwell equations and taking into account that $\mu_0 = \mu = 1$ (Gaussian units are used) the following set of equations is obtained:

$$E_z = -\frac{m}{\epsilon_3} H_y ,$$

$$H_z = mE_y ,$$

$$H_y = -i\frac{c}{\omega} \left[1 - \frac{m^2}{\epsilon_3} \right]^{-1} \frac{dE_x}{dz} ,$$

(2.4)

$$H_x = i\frac{c}{\omega} \frac{dE_y}{dz} ,$$

whereas E_x, E_y are related by the coupled equations

$$\left[\frac{1}{\epsilon_m} \frac{c^2}{\omega^2} \frac{d^2}{dz^2} + 1 - \frac{m^2}{\epsilon_3} \right] E_x + \delta \left[1 - \frac{m^2}{\epsilon_3} \right] (E_x\cos2qz + E_y\sin2qz) = 0 ,$$

$$\left[\frac{1}{\epsilon_m} \frac{c^2}{\omega^2} \frac{d^2}{dz^2} + 1 - \frac{m^2}{\epsilon_m} \right] E_y + \delta(E_x\sin2qz - E_y\cos2qz) = 0 .$$

(2.5)

This set of equations is equivalent to the one reported in Refs. 1 and 4—6. The periodicity of the structure along z, with period $p/2 = 2\pi/2q$, suggests looking for solutions of the form

$$\begin{bmatrix} E_x \\ iE_y \end{bmatrix} = e^{iK_\parallel z} \sum_{n=-\infty}^{\infty} \begin{bmatrix} x_n \\ y_n \end{bmatrix} e^{2niqz} .$$

(2.6)

According to Floquet's theorem the set of Eqs. (2.5) has at least one solution of the type (2.6). Generally, how-

ever, there exists a complete set of solutions of this type, except in special cases which will be discussed in the following. For a more complete discussion the reader should refer to the paper by Dreher et $al.$[5]

Substituting Eq. (2.6) into Eq. (2.5), the following three-diagonal set of algebraic equations is obtained:

$$b_{n-1}u_{n-1}+a_{n-1}v_{n-1}+c_{n-1}u_n = 0$$
$$c_n v_{n-1}+a_n u_n+b_n v_n = 0$$
$$b_n u_n+a_n v_n+c_n u_{n+1} = 0 .$$

$$\therefore$$

(2.7)

Here

$$u_n = x_n - y_n, \quad v_n = x_n + y_n ,$$ (2.8)

and the meaning of the other variables is given in the following list:

$$a_n = a - a'(k+n)^2 ,$$ (2.9)

$$b_n = b - b'(k+n)^2 ,$$ (2.10)

$$c_n \equiv c = \epsilon_m \delta m' \omega_r^2 ,$$ (2.11)

$$a = \frac{m'}{2}(2\epsilon_m - m^2)\omega_r^2 ,$$ (2.12)

$$b = \frac{m'}{2}m^2\omega_r^2 ,$$ (2.13)

$$a' = \frac{1}{2}\left[\frac{1}{m'}+m'\right] ,$$ (2.14)

$$b' = \frac{1}{2}\left[\frac{1}{m'}-m'\right] ,$$ (2.15)

$$m' = \left[1-\frac{m^2}{\epsilon_3}\right]^{1/2} ,$$ (2.16)

$$m = \frac{K_\perp c}{\omega} ,$$ (2.17)

$$\epsilon_m = \frac{\epsilon_1+\epsilon_2}{2}, \quad \delta = \frac{\epsilon_1-\epsilon_2}{\epsilon_1+\epsilon_2} ;$$ (2.18)

$$q = \frac{2\pi}{p}, \quad p = \text{helix pitch} ;$$ (2.19)

$$\omega_r = \frac{\omega}{2qc} = \frac{p}{2\lambda} ,$$ (2.20)

$$k = \frac{K_\parallel}{2q} .$$ (2.21)

III. EVALUATION OF THE COEFFICIENTS DETERMINANT

Equations (2.7) represent a system of infinitely many homogeneous equations in the unknown quantities $\{u_n, v_n\}$. The coefficient determinant is a continuant and will be indicated by the following symbol:

$$\begin{vmatrix} \cdots & b_{n-1} & c_{n-1} & b_n & c_n & \cdots \\ \cdots & a_{n-1} & a_n & a_n & \cdots \\ \cdots & b_{n-1} & c_n & b_n & c_{n+1} & \cdots \end{vmatrix} .$$ (3.1)

Here the second row is formed by the elements of the main diagonal and the other two rows by the elements of the upper and lower diagonal, respectively.

Such a determinant is obviously divergent, due to the fact that its elements grow as n^2 for large values of n.

In order to obtain a set of equations having a convergent determinant, it is convenient to divide both sides of (2.7) by $n^2 - k_0^2$, where k_0 is a suitable chosen constant. The corresponding determinant will be considered as a function of k and indicated by $D(k)$, i.e.,

$$D(k) = \begin{vmatrix} \cdots & \cdots & \frac{c_{n-1}}{(n-1)^2-k_0^2} & \frac{b_n}{n^2-k_0^2} & \frac{c_n}{n^2-k_0^2} & \cdots \\ \cdots & \frac{a_{n-1}}{(n-1)^2-k_0^2} & \frac{a_n}{n^2-k_0^2} & \frac{a_n}{n^2-k_0^2} & \cdots \\ \cdots & \frac{b_{n-1}}{(n-1)^2-k_0^2} & \frac{c_n}{n^2-k_0^2} & \frac{b_n}{n^2-k_0^2} & \cdots & \cdots \end{vmatrix} .$$ (3.2)

With the use of a method similar to the one used for the Hill determinant,[7] it is possible to represent $D(k)$ in the form of a simple analytic function of k. We divide first the rows of $D(k)$ by $[(k+n^2)-k_0^2]/(n^2-k_0^2)$ and write

$$D(k)= \prod_{n=-\infty}^{+\infty} \left[\frac{(k+n)^2-k_0^2}{n^2-k_0^2} \right]^2 D'(k) , \tag{3.3}$$

where

$$D'(k)= \begin{vmatrix} \cdots & \cdots & \gamma_{n-1} & \beta_n & \gamma_n & \cdots \\ \cdots & \alpha_{n-1} & \alpha_n & \alpha_n & \cdots \\ \cdots & \beta_{n-1} & \gamma_n & \beta_n & \cdots & \cdots \end{vmatrix} \tag{3.4}$$

with

$$\alpha_n = \frac{a-a'(k+n)^2}{(k+n)^2-k_0^2} , \tag{3.5}$$

$$\beta_n = \frac{b-b'(k+n)^2}{(k+n)^2-k_0^2} , \tag{3.6}$$

$$\gamma_n = \frac{c}{(k+n)^2-k_0^2} . \tag{3.7}$$

It is straightforward to check that $D'(k)$ has the following properties:

(1) $D'(k)=D'(k+n)$,   (3.8)

(2) $D'(k)=D'(-k)$,   (3.9)

and (3) $D'(k)$ is an analytic function of k with second-order poles at points represented by the two progressions

$$k'_n = k_0+n , \tag{3.10}$$

$$k''_n = -k_0+n , \tag{3.11}$$

where n is any integer.

Let $C_1(k)$ and $C_2(k)$ be the coefficients of the dipole and quadrupole terms of $D'(k)$ corresponding to a given singular point k_n belonging to one of these progressions. Then, according to Eq. (3.8) all the poles belonging to the same progressions have the same coefficients,

$$C_1(k)=C_1(k+n)=C_1 , \tag{3.12}$$

$$C_2(k)=C_2(k+n)=C_2 . \tag{3.13}$$

Furthermore, according to Eq. (3.9), all the poles belonging to different progressions have the same coefficient C_2 and opposite coefficient C_1,

$$C_1(k)=-C_1(-k)=C_1 , \tag{3.14}$$

$$C_2(k)=C_2(-k)=C_2 . \tag{3.15}$$

The function

$$f(k)=D'(k)-\sum_{-\infty}^{\infty} \left[\frac{C_1}{k-k_0-n} - \frac{C_1}{k+k_0-n} + \frac{C_2}{(k-k_0-n)^2} + \frac{C_2}{(k+k_0-n)^2} \right] \tag{3.16}$$

is therefore analytic, with no singularities, and bounded. By the Liouville theorem it is then a constant, which in the following will be indicated by the symbol C_0.

The constant C_2 is given by

$$C_2 = \lim_{k\to k_0} (k-k_0)^2 D'(k) , \tag{3.17}$$

whereas C_0 and C_1 can be obtained once $D'(k)$ is known for two different values of k. Let these values be $k=0$ and $\frac{1}{2}$. Then from the relations

$$\sum_{n=-\infty}^{\infty} \frac{1}{(z-n)^2} = \frac{\pi^2}{\sin^2 \pi z} , \tag{3.18}$$

$$\sum_{n=-\infty}^{\infty} \frac{1}{z-n} = \pi \cot \pi z , \tag{3.19}$$

we easily obtain

$$C_0 = \frac{D'(0)\sin^2\pi k_0 + D'(\frac{1}{2})\cos^2\pi k_0 - 4\pi^2 C_2}{\sin^2\pi k_0 + \cos^2\pi k_0} , \qquad (3.20)$$

$$C_1 = \frac{D'(0) - D'(\frac{1}{2}) - 2\pi^2 C_2(1/\sin^2\pi k_0 - 1/\cos^2\pi k_0)}{1/\sin^2\pi k_0 + 1/\cos^2\pi k_0} . \qquad (3.21)$$

Recalling that

$$\prod_{n=1}^{\infty} \left[1 - \frac{z^2}{n^2} \right] = \frac{\sin\pi z}{\pi z} , \qquad (3.22)$$

we finally obtain

$$D(k) = \frac{C_0}{\sin^4\pi k_0}(\sin^4\pi k - 2U\sin^2\pi k + V) , \qquad (3.23)$$

where

$$U = \sin^2\pi k_0 + \frac{C_1}{2C_0} - \frac{\pi^2 C_2}{C_0}(1 - 2\sin^2\pi k_0) , \qquad (3.24)$$

$$V = \sin^4\pi k_0 + \frac{C_1 + 2\pi^2 C_2}{C_0}\sin^2\pi k_0 . \qquad (3.25)$$

In order to actually obtain $D(k)$ it is thus necessary only to know $D'(0)$, $D'(\frac{1}{2})$, and C_2, which implies that it is necessary to evaluate the continuant for three values of k. This can be performed numerically on a suitably truncated form of the continuant. The parameter k_0—yet to be assigned—can be chosen in such a way as to minimize the truncation errors.

In order to make the above choice, it should be noted that the elements γ_n of the continuant tend to approach zero as $1/n^2$ and that if a term contains γ_n, it contains γ_{n-1} as well. Thus for $n \to \infty$ these terms tend to approach zero as $1/n^4$ and become negligible for sufficiently large value of n. When this happens, the continuant reduces to a product of factors of the form

$$\begin{vmatrix} \alpha_n & \beta_n \\ \beta_n & \alpha_n \end{vmatrix} = \frac{[(n+k)^2 - k_a^2][(n+k)^2 - k_b^2]}{[(n+k)^2 - (k_a^2 + k_b^2)/2]^2} , \qquad (3.26)$$

where

$$k_a = (\epsilon_m)^{1/2}m'\omega_r , $$
$$k_b = (\epsilon_m - m^2)^{1/2}\omega_r . \qquad (3.27)$$

In order to have a rapid convergence of the continuant it is thus convenient to set

$$k_0 = \frac{1}{\sqrt{2}}(k_a^2 + k_b^2)^{1/2} . \qquad (3.28)$$

An even quicker convergence can be obtained by writing in a slightly different way the basic equations. Instead of dividing by $(n^2 - k_0^2)$ the two equations of the system (2.7) containing n^2, the first equation is divided by $(n^2 - k_a^2)$ and the second by $(n^2 - k_b^2)$. It is thus possible to obtain an equation similar to (3.23), where $D(k)$ is still proportional to $(\sin^4\pi k - 2U\sin^2\pi k + V)$, but the quantities U and V are given by the following expressions:

$$2U = \sin^2\pi k_a + \sin^2\pi k_b - C_a - C_b , \qquad (3.29)$$

$$V = \sin^2\pi k_a\sin^2\pi k_b - C_a\sin^2\pi k_b - C_b\sin^2\pi k_a , \qquad (3.30)$$

where

$$C_a = \pi\sin(2\pi k_a)\lim_{k\to k_a}(k - k_a)D^*(k) , \qquad (3.31)$$

$$C_b = \pi\sin(2\pi k_b)\lim_{k\to k_b}(k - k_b)D^*(k) , \qquad (3.32)$$

and $D^*(k)$ is a new determinant, given by

$$D^*(k) = \begin{vmatrix} \cdots & & \frac{c}{(k+n-1)^2 - k_b^2} & \frac{b - b'(k+n)^2}{(k+n)^2 - k_a^2} & & \frac{c}{(k+n)^2 - k_b^2} & \cdots \\ & \frac{a - a'(k+n-1)^2}{(k+n-1)^2 - k_b^2} & & \frac{a - a'(k+n)^2}{(k+n)^2 - k_a^2} & & \frac{a - a'(k+n)^2}{(k+n)^2 - k_b^2} & \cdots \\ \cdots & & \frac{c}{(k+n)^2 - k_a^2} & \frac{b - b'(k+n)^2}{(k+n)^2 - k_b^2} & & \frac{c}{(k+n+1)^2 - k_a^2} & \cdots \end{vmatrix} . \qquad (3.33)$$

The advantage of $D^*(k)$ with respect to $D'(k)$ is that the sum of all the terms which do not contain the constant c is equal to 1.

In order to give an idea of the rapidity of convergence of these continuants, we note that for all the numerical calculations reported in this paper it was never necessary to take into account terms with $|n| > 15$ to reduce the truncation error to less than 1 ppm.

IV. CHARACTERISTIC EQUATION

On the basis of the results of Sec. III, the characteristic equation reads as

$$\sin^4\pi k - 2U(\omega,m)\sin^2\pi k + V(\omega,m) = 0 \ . \qquad (4.1)$$

The quantity m appearing in U and V is, according to Eq. (2.4), proportional to $\sin\theta_i$, where θ_i is the incidence angle. It is worth noting that, if k is a solution of Eq. (4.1), $-k$ and $k+n$ are solutions as well.

The simplicity of this equation suggests that a simpler way to obtain it should exist. It is actually easy to show, using a method very similar to the one used by Dreher $et \ al.$, that the characteristic equation has to be a quadratic equation in the variable $\sin^2\pi k$. The present method however allows the direct calculation of the coefficients U and V.

Equation (4.1) has four roots for $\sin\pi k$ and thus four sets of characteristic values for k,

$$k_1^+ = k_1 + n, \quad k_2^+ = k_2 + n,$$
$$k_1^- = -k_1 + n, \quad k_2^- = -k_2 + n \ . \qquad (4.2)$$

For each set, Eqs. (2.7) have unique independent solution. Indeed, if different values of k belonging to the same set are substituted in Eq. (2.7), only a reindexing of the components $\{x_n, y_n\}$ occurs.

It should be noted that the two solutions of the basic equations (2.7) corresponding to k_i^+ and k_i^- represent waves that propagate in opposite directions along the z axis. Thus for any point of the plane (ω, m) there exist only two really independent modes.

Such solutions have very different characteristics, depending on the particular values of k. The following three cases should be distinguished.

(A) $\sin^2(\pi k)$ is real and its value is between zero and one. The exponent k is then real and the solution is stable.

(B) $\sin^2(\pi k)$ is real and its value is larger than 1 or smaller than zero. The exponent k is then complex and its real part is an integer or a half-integer, respectively. The real part of k satisfies the Bragg condition $\mathrm{Re}k = \frac{1}{2}n$, n being even or odd depending on the case. The corresponding solutions of Eqs. (2.7) are unstable and give rise to Bragg reflection peaks.

(C) $\sin^2(\pi k)$ is complex. In this case also k is complex and its real part does not satisfy the Bragg condition. The corresponding solutions are unstable and, for a semi-infinite sample, produce total reflection, as in case (B). This type of instability can arise in two different situations. The first one corresponds to the case where the incidence angle θ_i is small and the discriminant $U^2 - V$ of Eqs. (4.1) is negative, while the second one occurs when $m = n_i\sin\theta_i$ is sufficiently large so that, according to Eqs. (2.9)—(2.17), the coefficients a_n, b_n, c_n of Eq. (2.7) become complex, and thus also the coefficients U and V may be complex.

On the separation lines between stability and instability regions, the characteristic equation gives degenerated roots. In this case a basic set of solutions of Eqs. (2.5), satisfying the Bloch-Foquet theorem, no longer exists.
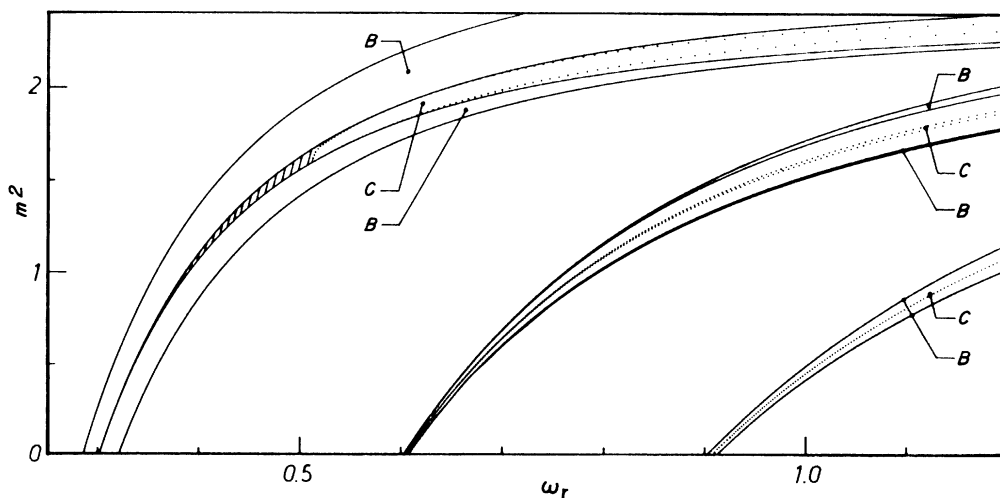
In Fig. 1 a typical "chart of stability" is reported,

FIG. 1. Chart of stability computed for $\epsilon_1 = 3.06$ and $\epsilon_2 = \epsilon_3 = 2.43$. Solid lines are the loci of the points where one of the internal modes of propagation satisfies the Bragg condition; they are at the boundaries of the instability regions of type B. Dotted lines are the loci of the points where the discriminant of the characteristic equation vanishes so that the two internal modes reduce to only one; they are the boundaries of the instability regions of type C. ω_r is the reduced frequency, m is a quantity proportional to $\sin\theta_i$, where θ_i is the incidence angle. In the shaded region both modes are of type B.
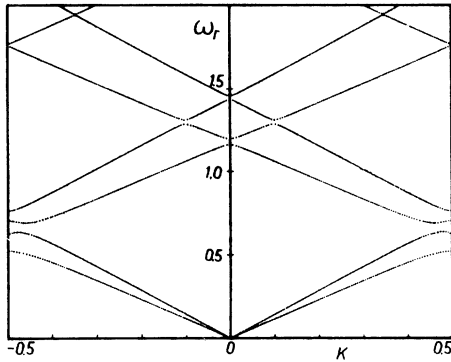
FIG. 2. Dispersion curves for the two elementary modes in the reduced Brillouin zone, computed at fixed incidence angle for the cholesteric of Fig. 1 and for $m = \sqrt{2}$. Frequency gaps which occur at $k = \pm 0.5$ and at $k = 0$ correspond to instability regions of type B; they give rise to the Bragg reflection peaks. Other frequency gaps, which are common to both modes, correspond to instability regions of type C; they give rise to further reflection peaks between two Bragg peaks.

in which regions of instability of type B and C are shown. This chart is similar to the one appearing in Dreher's paper but shows also the existence of regions of type C which do not appear in Dreher's plots. Such regions are included within the two Bragg bands corresponding to the two elementary modes.

The instability regions correspond to total reflection bands. It can be noted that all the bands of order $n > 1$ becomes vanishingly narrow for $m \rightarrow 0$. This agrees with the well-known result that for normal incidence only a single reflection band exists.

Finally, in Figs. 2 and 3 are reported the dispersion curves $\omega_r(k)$ and $k(\omega_r)$ for $m^2 = 2$. It can be observed that the curves represented in the reduced
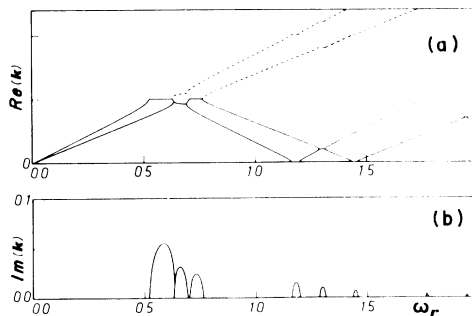

FIG. 3. Real part Rek and imaginary part Imk of the reduced wave vector k vs ω_r, computed in the same conditions of Fig. 2. Rek solid lines represent the dispersion curves in the reduced Brillouin zone. $2q$ Imk is the attenuation constant of the mode, its inverse is the amplitude attenuation length.

Brillouin zone (solid lines) cross exactly at the boundaries of the type-C region.

The detailed study of the solutions of the basic equations (2.7) is still in progress and will be published in a subsequent paper.

V. DISCUSSION

The curves reported in Figs. 1—5 refer to the cholesteric liquid crystal studied by Berreman and Scheffer[6] which is a binary mixture of nonmesomorphic dextro 4,4'-bis(2-methylbutoxy) azoxybenzene (2MBAB) and nematic 4,4'-bis(hexyloxy) azoxybenzene, with 0.45 molar fraction of 2MBAB. As reported by these authors, this cholesteric has a pitch of 0.764 μm at 88 °C and the following values of the dielectric tensor: $\epsilon_1 = 3.060$ and $\epsilon_2 = \epsilon_3 = 2.430$. These values have been obtained by comparing the experimental reflectance from a single-domain thin film with that calculated by the method of propagation matrices.

Some disagreements between experiments and theoretical results are pointed out by the authors quoted above. We found it interesting to carry out the calculations by a different method, also because we found it puzzling that in their theoretical curves the second-order Bragg reflection band shows essentially only two peaks. As also in the paper of Dreher and Meyer the presence of the central peak
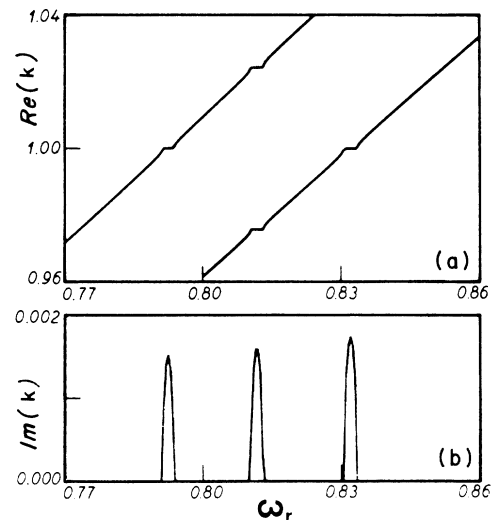

FIG. 4. Real part Rek and imaginary part Imk of the reduced wave vector k computed for the cholesteric of Fig. 1 and for $m = 1.06$, corresponding to light incident at 45° from the glass, around the second-order reflection band. Curves show that this reflection band is a triplet and that the three components have comparable heights and widths. Ratios of the areas of the three Im(k) peaks are 1:1.2:1.4.

is ignored; one might doubt the very existence of such a peak.

We therefore calculated the reflectance of a flat layer around the second-order band, assuming the same conditions of Ref. 6, i.e., for light incident at 45° in the x,y plane on a sample of 15 pitch lengths. The calculation goes through the following steps.

(a) By means of Eq. (4.1) the characteristic values are computed. In Fig. 4 the real and imaginary parts of k are reported. The quantity $2q \, \text{Im}(k)$ is the attenuation constant. One can notice three well-separated instability zones of comparable width and height. The energy density attenuation length at the center of these zones is about 25 pitches. Because the sample was 15 pitch lengths, we expect three accentuated reflectance peaks of comparable intensity.

(b) By Eq. (2.7) the coefficients u_n and v_n, which define the spectral composition of each mode, are computed. These coefficients vanish very rapidly as n increases. For such a reason they can be obtained with any desired approximation by omitting all u_n, v_n for $n > n_0$, where n_0 is some positive integer. The obtained results can be summarized as follows. The mode which corresponds to the upper curve of Fig. 4 is elliptically polarized with a very strong E_y component (E_x is about one tenth of E_y). The oppo-

site occurs for the other mode. A different situation occurs only in a very narrow interval inside and around the C-type instability zone, where both modes have E_x and E_y components of comparable intensity.

(c) Finally, when the four elementary modes of characteristic values $k_1^+, k_2^+, k_1^-, k_2^-$ are known, the wave inside the sample is represented as a linear combination of them. The four coefficients of the linear combination appear as unknown quantities in a system of eight linear equations, obtained by matching the fields at the layer surfaces. The other four unknowns are the E_x and E_y components of the transmitted and reflected waves. The cases of π and σ polarization of the incident light have been separately considered.

In Fig. 5 the reflectance coefficients versus ω_r are reported. We note that each one of the four $R_{\pi\pi}$, $R_{\sigma\sigma}$, $R_{\pi\sigma}$, and $R_{\sigma\pi}$ curves shows only one peak well above the interference fringes. The peaks of $R_{\pi\pi}$ and $R_{\sigma\sigma}$ curves are the Bragg peaks of each of the two modes. The peaks of $R_{\pi\sigma}$ and $R_{\sigma\pi}$ are instead related to the C-type instability. This means that the Bragg reflection is nearly in the same polarization state of the incident beam, while the reflection from C-type instability is prevalently polarized in the opposite state. Our calculations yield results practically coincident with the theoretical curves of Ref. 6 which, however, reports only the $R_{\pi\pi}$ and $R_{\sigma\sigma}$ curves, where the C-type reflection peak is nearly absent. The disagreements between theory and experiment are confirmed.

It must be noted, anyhow, that all the calculations have been made assuming a perfectly ordered sample, while the real sample is certainly somewhat diffusing owing to surface defects and thermal fluctuations of the director. At present no reliable calculation has been published on the light scattering properties of cholesteric liquid crystals. The present theory has been developed also in order to study the light scattering of thermally excited internal modes. Taking into account that for a nematic liquid crystal with the same thickness and optical anisotropy the scattered intensity is about 10% of the incident light (Ref. 8), a non-negligible effect on the reflectance properties of a cholesteric sample should be expected.

FIG. 5. Second-order reflectance spectra of a cholesteric liquid-crystal film 15 pitch lengths or 11.45-μm thick in the same conditions of Fig. 4. Indices π and σ denote the polarization of the incident and reflected light with respect to the incidence plane: π indicates parallel and σ indicates normal.

VI. CONCLUSIONS

Despite the fact that the interest in cholesteric liquid crystals lies in their extraordinary optical properties, astonishingly enough, these properties are far from being completely understood. Most of the papers found in the literature are concerned with the particular case of light propagating along the helical axis. The more general case of oblique prop-
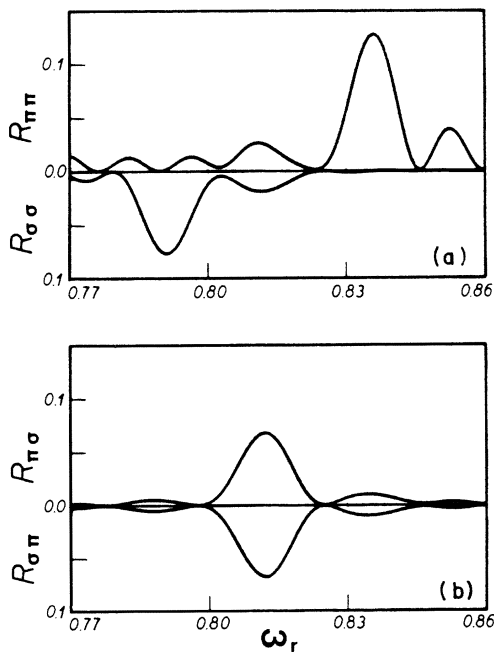
agation is usually treated with numerical methods. These methods are useful for studying some particular properties of these liquid crystals, but they give little insight on the light-propagation mechanism.

In the present paper a new formalism has been introduced which is similar to the one used to characterize the light propagation in the homogeneous, optically anisotropic crystals. That is, we consider the wave inside the cholesteric as a superposition of plane waves having a definite polarization state. The method is not very different from that used by Taupin to calculate the reflectance from a semi-infinite sample and from that used by Dreher and Meier to find the Bragg reflection bands. In the present paper, however, the formalism is developed to the point to make it applicable to study the general optical properties of helicoidal structures, as, for instance, the reflectance and the transmittance of cholesteric liquid crystal films. The actual calculations are carried out on the basis of reasonably simple expressions and yield very precise numerical results.

Among the preliminary results thus obtained we would like to stress what follows. We obtained a very simple equation for the characteristic vectors. The very existence of such an equation allows us to classify the different types of instabilities occuring in a helicoidal medium. We were then able to clarify one of the most controversial points in the literature, i.e., the number and the origin of the reflection peaks. We show that the stability properties of the electromagnetic wave is determined by the coexistence of two modes with different polarization states. Between the Bragg reflection peaks of the same order of the two modes, a further peak is evidenced, where the dispersion curves in the first Brillouin zone intersect. This peak is common to both modes and is therefore present independently of the polarization state of the incident light, while the Bragg instabilities give rise to selective reflection. For what concerns the polarization of the reflected light, we note that the Bragg reflection peaks are predominating of the $\pi\pi$ or $\sigma\sigma$ type, while in the central peak the dominant component is of the $\pi\sigma$ or $\sigma\pi$ type.

ACKNOWLEDGMENTS

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