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Structure of reflection bands of Grandjean textures

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The characteristic exponents of the normal modes for light in the Grandjean texture of a cholesteric film were calculated with values for the dielectric constants that are typical for common cholesteryl derivatives. The reflection properties are discussed in terms of the penetration depth given by the real part of the characteristic exponent. The results are presented in graphs which give the structure and the reflection coefficients as a function of wavelength and angle of incidence. It was found that there are unpolarized bands that are missing in a stability chart published earlier.

I. INTRODUCTION

Cholesteric liquid crystals have a spontaneous twist that results in a helical structure. A typical texture of cholesteric films, known as Grandjean texture, is characterized by a perpendicular alignment of the twist axis. Its most striking optical properties are bands of selective reflection. The bands have been studied in a number of theoretical and experimental papers (for reviews, see Ref. 1). The reflection is due to internal interference. There is a sequence of reflection regions corresponding to different interference orders. Accurate solutions of the wave equation can be given in closed form only for perpendicular incidence. For oblique incidence approximate expressions or numerical approaches must be used.[2-6] Taupin[2] was the first to notice on the basis of theoretical studies that, at least for sufficiently oblique incidence, a triplet of bands belongs to each reflection order. Theoretical and experimental studies at an angle of incidence of 45° have been made by Berreman and Scheffer.[3] The agreement between experiment and theory is good. The observed second-order reflection consists clearly of a triplet of separate bands. The first-order reflection is strongly structured but not separated into a triplet. For vertical incidence only the first-order reflection is observed and, in agreement with theoretical predictions, it consists of a single band without structure.[7]

A general survey of the dependence of the reflection bands on wavelength and angle of incidence is presented in the stability chart by Dreher and Meier.[5] The chart predicts, in contrast to other results, a doublet of bands for each reflection order at oblique incidence, and therefore it is necessary to recheck the derivations.

We extend in this paper the earlier calculations[5] and confirm the existence of triplets of bands. The two outer bands of each triplet give selective reflection of polarized light, while the center band is unpolarized. The first-order reflection shows a complex dependence on the angle of incidence. It changes from a single polarized band at vertical incidence to two overlapping polarized bands at oblique incidence, and only at very oblique incidence it assumes the described triplet structure. We recollect and summarize in Sec. II relevant points of the general theory and clarify the cause of the discrepancy. New results of numerical calculations are presented and discussed in Secs. III and IV.

II. CHARACTERISTIC EXPONENTS AND PENETRATION DEPTH

In the Cartesian coordinate system with the z axis parallel to the twist axis, the dielectric tensor for the Grandjean texture may be written in the form

$$\begin{aligned}
&\epsilon_{xx}=\bar{\epsilon}+(\epsilon_a/2)\cos qz,\\
&\epsilon_{yy}=\bar{\epsilon}-(\epsilon_a/2)\cos qz,\\
&\epsilon_{zz}=\epsilon_2,\\
&\epsilon_{xy}=\epsilon_{yx}=(\epsilon_a/2)\sin qz,\\
&\epsilon_{xz}=\epsilon_{zx}=\epsilon_{zy}=\epsilon_{yx}=0.
\end{aligned}\tag{1}$$

We assumed that the local dielectric tensor has rotational symmetry and set

$$\bar{\epsilon}=(\epsilon_1+\epsilon_2)/2,$$

$$\epsilon_a=\epsilon_1-\epsilon_2,$$

where ϵ_1 and $\epsilon_2 = \epsilon_3$ are the principal dielectric constants, and $q = 4\pi/p$, where p denotes the pitch, i.e., the distance between planes of the same director orientation.

We are interested in modes that can be excited by incoming plane waves that propagate in the x,z plane. These modes are obtained by superposition of normal modes of the form[3]

$$\vec{E} = \vec{F}(z) e^{i(\omega t - k_x x)} \tag{2}$$

with $k_x = n_{\text{ex}}(2\pi/\lambda)\cos\gamma$, where n_{ex} denotes the external refractive index, γ the angle of incidence, and λ the vacuum wavelength. $\vec{F}(z)$ satisfies the differential equations

$$\begin{aligned} \frac{d^2F_x}{dz^2} &= \left[\frac{k_x^2}{\epsilon_2} - \frac{\omega^2}{c^2} \right] \\ &\quad \times \left[\bar{\epsilon} F_x + \frac{\epsilon_a}{2} (F_x \cos qz + F_y \sin qz) \right], \\ \frac{d^2F_y}{dz^2} &= \left[\frac{k_x^2}{\bar{\epsilon}} - \frac{\omega^2}{c^2} \right] \bar{\epsilon} F_y \\ &\quad + \frac{\epsilon_a}{2} \frac{\omega^2}{c^2} (F_y \cos qz - F_x \sin qz), \\ F_z &= \frac{ik_x}{k_x^2 - \frac{\omega^2}{c^2}\epsilon_2} \frac{dF_x}{dz} \; . \end{aligned} \tag{3}$$

The equations have four independent solutions, and with the exception of singular lines in the γ,λ plane all four have the form

$$\vec{F} = e^{-\mu z} \sum_{\nu=-\infty}^{\infty} A_\nu e^{i\nu qz}, \tag{4}$$

where $\mu = q(\alpha + i\beta)$ is the characteristic exponent. β is, of course, determined only up to an additive integer. For uniqueness we require $-\frac{1}{2} < \beta \leq \frac{1}{2}$. It can be seen that the solutions of (3) occur in pairs, i.e., to each solution $\vec{F}(z) = (F_x(z), F_y(z), F_z(z))$ and the characteristic exponent μ_F, there exists a second solution $\vec{G}(z) = (-F_x(-z), F_y(-z), F_z(-z))$ with $\mu_G = -\mu_F$. The second solution corresponds to the reflected mode. The z component of its Poynting vector has the opposite sign. The forward and reflected modes have, with the exception of the sign, the same characteristic exponent so that for each k_x only two independent exponents and two independent basic modes, which we denote in the following as modes (1) and (2), exist.

In the absence of absorption there exist additional relations between the exponents. In that case all ϵ_{ik} are real, and therefore with μ the conjugate complex μ^* is also a characteristic exponent. In sum this gives the following set of related exponents:

$$\alpha + i\beta, \; \alpha - i\beta, \; -\alpha + i\beta, \; -\alpha - i\beta. \tag{5}$$

They are usually pairwise equivalent, i.e., when $\alpha = 0$ or when $\beta = 0$ or $\frac{1}{2}$.

In addition, the following general statements can be made on the form of the exponents of the two modes:

(a) Outside the reflection regions $\alpha = 0$ and

$$\mu_{(1)} = \pm iq\beta_1, \quad \mu_{(2)} = \pm iq\beta_2; \tag{6}$$

(b) regions with one reflection band

$$\mu_{(1)} = \pm q\alpha_1 + in_1 q/2, \quad \mu_{(2)} = \pm iq\beta_2; \tag{7}$$

or

$$\mu_{(1)} = \pm iq\beta_1, \quad \mu_{(2)} = \pm q\alpha_2 + in_2 q/2 \; ; \tag{7$'$}$$

(c) regions with overlapping reflection bands. Here two significantly different possibilities exist. The first one is

$$\begin{aligned} \mu_{(1)} &= \pm q\alpha_1 + in_1 q/2, \\ \mu_{(2)} &= \pm q\alpha_2 + in_2 q/2. \end{aligned} \tag{8}$$

The second possibility is

$$\mu_{(1)} = \pm q(\alpha_1 + i\beta_1), \quad \mu_{(2)} = \mu^*_{(1)}. \tag{9}$$

In the sets (7) and (8) n_1 and n_2 are independently either 0 or 1 so that for all sets except for (9), $\mu^*_{(j)} = -\mu_{(j)}$ or $\mu^*_{(j)} = \mu_{(j)} - inq$.

We call reflection bands for which $\beta = 0$ or $\frac{1}{2}$ bands of the first kind and bands for which (9) holds of the second kind. Assuming μ to be a steady function of λ and γ the boundaries of reflection bands are given by the curves of double roots for μ. These are the singular lines mentioned before. Specifically, we have at the boundaries of bands of the first kind to regions without reflection for the exponent of the corresponding mode

$$\mu = inq/2, \quad n = 0 \text{ or } 1; \tag{10}$$

at the double boundaries of bands of the second kind to regions without reflection

$$\mu_{(1)} = \mu_{(2)} = \pm iq\beta, \tag{11}$$

and at the double boundaries between a band of the second kind to a region with overlapping bands of the first kind

$$\mu_{(1)} = \mu_{(2)} = \pm q\alpha + inq/2, \quad n = 0 \quad \text{or} \quad 1 \; . \tag{12}$$

The stability chart of the earlier study[5] contains only the curves $\mu = 0$ and $iq/2$. Pairs of curves were assigned as boundaries to reflection bands in a self-consistent way and so that the case of vertical incidence is given properly. The chart is incomplete because bands of the second kind occur and the boundaries given by (11) and (12) must also be included. In the present study the characteristic exponent is calculated quasicontinuously as a function of the angle of incidence, and uncertainties concerning the assignment of boundaries to reflection bands are avoided.

III. NUMERICAL RESULTS

It can be deduced from Eq. (3) that the reduced exponent $\mu/q=\alpha+i\beta$ depends on three dimensionless parameters, e.g.,

$$k_x/q=n_{\text{ex}}(p/2\lambda)\cos\gamma,$$
$$(2\lambda/p)^2/\bar{\epsilon}, \tag{13}$$
$$\epsilon_a/\bar{\epsilon}.$$

We used, however, the natural parameters in the numerical calculations with the following specific values for the fixed parameter:

$$\bar{\epsilon}=2.27-0.0001i,\ \ \epsilon_a=0.16,$$
$$n_{\text{ex}}=1.522,\ \ p=0.5\mu.$$

The dielectric constants are the same as those used in some of the earlier calculations[5] and are typical for common cholesteryl derivatives. A small imaginary part has been added to $\bar{\epsilon}$ for technical reasons as explained below.

The exponents for vertical incidence $\gamma=90°$ are given by

$$(\alpha+i\beta)_{\pm}=i\left\{\frac{1}{4}+\bar{\epsilon}\left[\frac{\omega}{qc}\right]^2\left[1+4\frac{\bar{\epsilon}}{\epsilon_a^2}\left[\frac{qc}{\omega}\right]^2\right]\right\}^{1/2}-\frac{i}{2}. \tag{14}$$

At oblique incidence the exponents were calculated using the iterative process described previously.[8] Beginning from $\gamma=90°$, we decreased γ by small steps and extrapolated the exponents to obtain consistent starting values for each iteration. In this way two separate curves $\mu_{(1)}(\gamma)$, $\mu_{(2)}(\gamma)$ result for a given λ/p, starting at

$$\mu_{(1)}(90)=q(\alpha+i\beta)_+$$

and

$$\mu_{(2)}(90)=q(\alpha+i\beta)_-.$$

In the absence of absorption, that is, for real dielectric constants, the extrapolation procedures may fail to give separate curves since there are points where $\mu(\gamma)$ has unsteady derivatives and bifurcations. The inclusion of absorption leads to steady derivatives and to separate curves even over regions of bands of the second kind. In the calculations the increments of γ were automatically re-

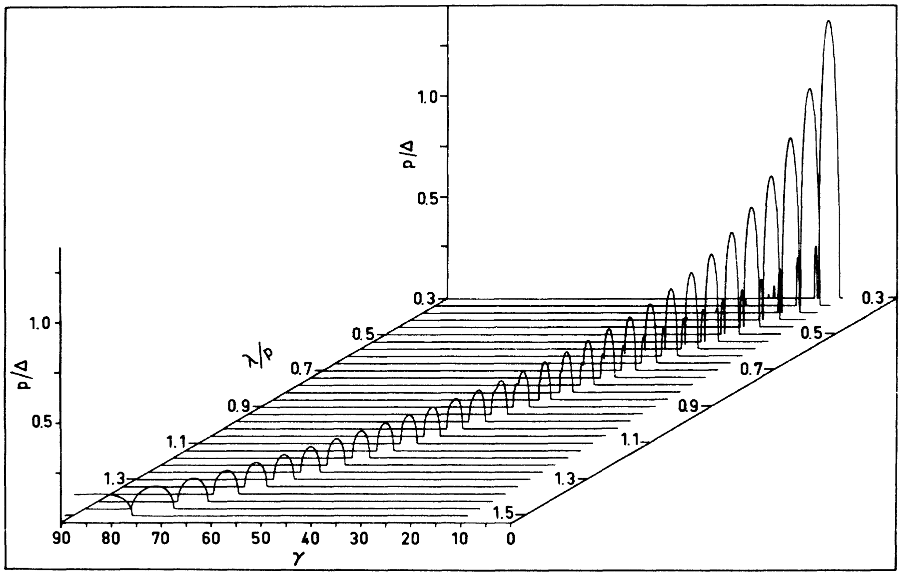

FIG. 1. Reduced reflection coefficients of mode (1) as function of angle of incidence and wavelength.

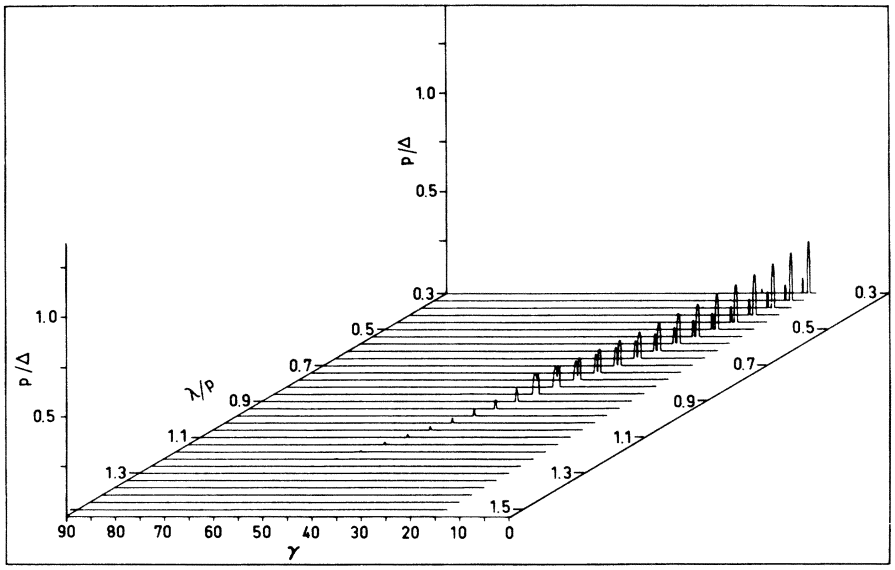

FIG. 2. Reduced reflection coefficients of mode (2) as function of angle of incidence and wavelength.

duced in the critical regions to a minimum of 0.001° from the normal value of 0.1°. For the magnitude of the imaginary part of $\bar{\epsilon}$ used in the calculations the rounding by absorption is small but clearly noticeable in Figs. 3 and 4. Figure 5 has been calculated with a ten times smaller imaginary part.

The real part of the characteristic exponent determines the reflection properties. We will refer to it in the following as a reflection coefficient. Its magnitude is equal to the inverse penetration depth $q\alpha = 1/\Delta$, where we define the penetration depth Δ as the distance from the interface at which the amplitude of an incoming mode has reduced to $1/e$ of its starting value. The results for the reduced reflection coefficients p/Δ are summarized in Figs. 1 and 2. Outside the reflection regions p/Δ is practically zero. Figure 1 demonstrates how the first-order reflection band of mode (1) shifts to smaller angles as the ratio λ/p decreases. At the same time the structure of the band changes. It begins to split and at small angles of incidence separates completely into two bands. For $\lambda/p = 1.54$ or larger ratios no selective reflection occurs.

The second normal mode (Fig. 2) has a first-order reflection band that reduces to zero width at vertical incidence. At small angles it also consists of two bands. One of these bands is of the second kind and shared with mode (1).

There is a sudden change of the structures of the reflection bands as obtained by our procedure in the range of λ/p near 0.88. Figures 3 and 4 show ex-

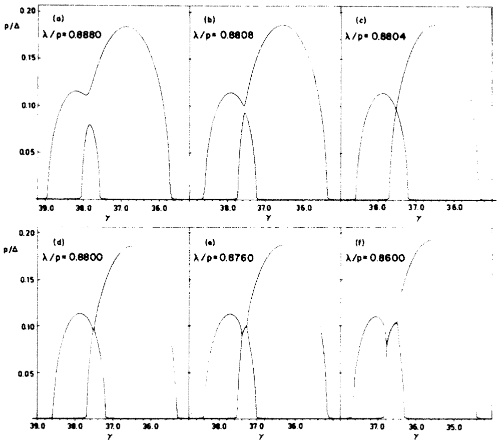

FIG. 3. Reduced reflection coefficients of modes (1) and (2) as functions of angles of incidence at indicated values of λ/p.

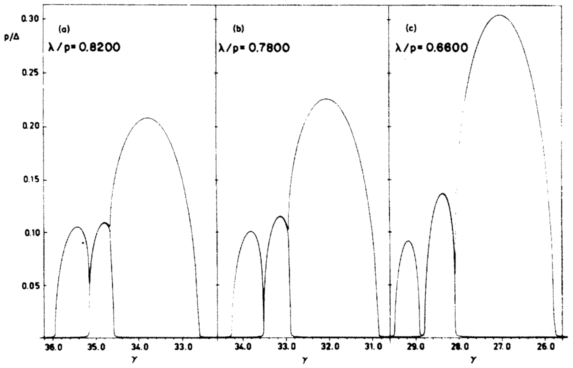

FIG. 4. Reduced reflection coefficients of modes (1) and (2) as functions of angles of incidence at indicated values of λ/p.

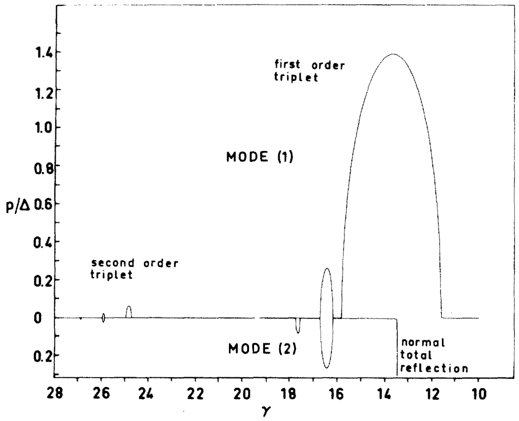

FIG. 5. Reduced reflection coefficients of modes (1) and (2) as functions of angles of incidence at $\lambda/p=0.3$. First- and second-order reflections are shown and the onset of the normal total reflection of mode (2) at $\gamma=13.46°$. The bands of the second kind are the same for both modes.

tended plots of the structures of the bands of modes (1) and (2). In Fig. 3(a) the two modes give overlapping bands of the first kind. The band of mode (1) begins to separate into a doublet. As λ/p decreases [Fig. 3(b)] the strength of band (2) increases, and at the same time the doublet character of band (1) becomes more pronounced. Finally, the discontinuous change in the band structures occurs near $\lambda/p=0.8804$ [Fig. 3(c)]. The maximum of band (2) reaches the minimum in the depression of band (1), and the curves can be reinterpreted so that a single reflection band is assigned to each mode. In this representation the shoulder of the original band of mode (1) is incorporated into the band of mode (2). With decreasing λ/p the band structures become again more complicated. A band of the second kind interferes [see Figs. 3(d)–3(f) and 4(a)–4(c)]. In the range of this band the two curves fall practically together. (The small deviations are due to absorption.) The band of the second kind becomes wider with decreasing λ/p ratio and finally forms a separate band.

In Fig. 5 the band of the second kind is completely separated. The first-order reflection ranges of modes (1) and (2) consist now of two bands each. Mode (2) reaches the angle of normal total reflection at $\gamma=13.48°$. The second-order reflections are also shown in Fig. 5. First- and second-order reflection bands have at this wavelength similar triplet structures with center bands that are of the second kind. Mode (1) has a relatively strong band of the first kind on the low angle side of the center band, while mode (2) has a weak band of the first kind on the side of higher angles of incidence.

In contrast to the first-order reflection, the second-order reflection remains a triplet of bands over the whole γ range. The width of these bands and the reflection coefficients reduces, of course, to zero as the reflection moves towards vertical incidence. Our calculations indicate also that other higher-order reflection bands have the same triplet structure. At very small angles of incidence and short wavelength the band structures are much more complicated since, as indicated in the stability chart,[5] bands of different order overlap.

IV. CONCLUSIONS

The results demonstrate that the earlier stability chart is incomplete because of the omission of the boundaries of the bands of the second kind. It is difficult to complement the chart because the missing boundaries are inconvenient to calculate. As far as the higher-order reflections are concerned only a center band has to be added, while more intricate changes are necessary for the first-order reflections.

The unpolarized bands are of particular interest. So far their existence has been clearly demonstrated only for the second-order reflection.[3] Good experimental evidence is still lacking for the first-order reflection. It has not been observed that the first order indeed separates into a triplet of bands at small angles of incidence.

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