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Arturo Mendoza, Eloy Muñoz-Pineda, Kenneth Järrendahl and Hans Arwin

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Evidence for a dispersion relation of optical modes in the cuticle of the scarab beetle Cotinis mutabilis

A. Mendoza-Galván,1,2* E. Muñoz-Pineda1,2, K. Järrendahl,2 and H. Arwin2,3

1Cinvestav-IPN, Unidad Querétaro, Libramiento Norponiente 2000, 76230 Querétaro, Mexico
2Laboratory of Applied Optics, Department of Physics, Chemistry and Biology, Linköping University, SE-581 83 Linköping, Sweden
3hans@ifm.liu.se

*amendoza@pravo.cinvestav.mx

Abstract: Variable angle Mueller matrix spectroscopic ellipsometry is used to study the properties of light reflected from the exoskeleton (cuticle) of the scarab beetle Cotinis mutabilis. For unpolarized incident light, the ellipticity and degree of polarization of the reflected light reveal a left-handed helical structure in the beetle cuticle. Analysis of the spectral position of the maxima and minima in the interference oscillations of the Mueller-matrix elements provides evidence for a dispersion relation similar to that of optical modes in chiral nematic liquid crystals calculated within a two-wave approximation. Additionally, a structural model for the cuticle of C. mutabilis is derived from the properties of the optical modes for non-attenuated propagation or selective reflection.

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References and links

1. Introduction

The properties shared between the cuticle (exoskeleton) of some species of beetles and chiral nematic (or cholesteric) liquid crystals (ChNLC) were identified some decades ago [1]: helical structure, large optical rotation, and reflection of near-circularly polarized light. Regarding the shared structure, in ChNLC systems rod-like molecules are preferentially oriented in pseudo-planes along a certain unit vector (the director). In the helical structure, the director of adjacent pseudo-planes is rotated a small constant angle about the helix axis. Depending upon the rotation direction, clockwise or counter-clockwise, the helical structure is right- or lefthanded. Additionally, single-domain structures are characterized by the presence of a single helical structure, otherwise the structure is of a multidomain type. The pitch of the structure corresponds to the distance separating two pseudo-planes when the director has completed a full rotation of 360°. In beetle cuticles, chitin-protein fibrils play the same role as rod-like molecules do in the ChNLC phase. Remarkably, electron microscopy images of beetle cuticles as well as of many other natural chiral systems show helical structures with the axis perpendicular to their surfaces [2]. From these observations it was hypothesized that the formation of single domain helical structures arises from an initial constraining layer [2]. Later, such hypothesis was qualitatively confirmed by numerical simulations using the Landau-de Gennes theory [3]. Owing to the helical structure of ChNLC and beetle cuticles, light incident at normal incidence is selectively reflected with the same handedness as that of the chiral structure. This type of selective reflection, commonly referred to as Bragg reflection, takes place in a spectral band centered at a wavelength equaling the product of the in-plane average refractive index and the pitch [1]. Even though reflection of light from beetle cuticles has been extensively studied over the past century, a renewed interest on the subject appeared during the latest decade. Thus, the comparison of experimental and simulated data at near-normal incidence using reflectance [4,5], ellipsometric [6], and Mueller-matrix spectra [7,8] have provided a qualitative description of beetle cuticles in terms of their thin-film analogue systems. In fact, a recent review shows how structures in living matter have inspired the improvement of optical performance of liquid crystals for the normal incidence case [9].

Bragg reflection at oblique incidence of single and multidomain ChNLC is also well understood from experimental and theoretical studies [10-13] as well as from calculations of the dispersion relation of the optical modes [14-17]. In contrast, research on the polarization properties of beetle cuticles at oblique incidence is at its beginning. Recently this issue has been addressed by our research group through the use of variable angle Mueller-matrix spectroscopic ellipsometry [18-22]. This multi-angle approach does not only account for the angle and spectral shift characteristic of all-dielectric reflective systems, but also for the appearance of interesting phenomena like the reflection of right-handed polarized light at increasing angles of incidence in some species [18,21]. Furthermore, structural parameters and refractive indices of beetle cuticles have been extracted by regression analysis of spectral multi-angle Muller-matrix data [19,20].

A close inspection of the available optical data from different species of beetle cuticles, at near-normal or oblique incidence, shows clear differences among the spectra. Some of them are rather smooth and broad, whereas modulation and interference oscillations are observed in data from other species. Particularly, in a previous work we reported the structure of the Mueller matrix of the scarab beetle Cotinis mutabilis (Gory and Percheron, 1833) and the presence of strong interference oscillations in the spectra was highlighted [21]. In this work, we focus on such interference oscillations with a twofold objective: i) to extract the structural and optical information therein contained and; ii) to investigate their relation with the polarization states of optical modes in ChNLC at oblique incidence. In order to achieve these objectives, this contribution is organized as follows. The experimental details and basics of Mueller matrices are presented in Sec. 2. In Sec. 3.1 the microstructure of the cuticle of C. mutabilis is briefly described. The polarization properties of reflected light from incident unpolarized light are discussed in Sec. 3.2. Evidence for a dispersion relation of optical modes in beetle cuticles is provided in Sec. 3.3 by analyzing the spectral dependence of maxima and minima appearing in the spectra of Mueller-matrix elements of C. mutabilis. This interpretation is supported in Sec. 3.4 with calculated dispersion relations for light propagation in cholesteric liquid crystals within a two-wave approximation [17]. In Sec. 3.5 a structural model for beetle cuticle is proposed on the basis of features in the experimental data and theoretical considerations for light propagation. Finally, the conclusions of this work are summarized.

2. Experimental

The beetle C. mutabilis is found in Mexico and the southwestern part of the United States [23]. It presents a green but not shiny color on its dorsal side with orange-brown stripes in the elytra. Its abdominal side presents a segmented structure and shows a shiny metallic-like color. The specimen under study was collected at Querétaro, Mexico. For the study, areas as flat as possible were selected from the segments in the abdomen and a small piece of about 2×2 mm² was cut using a sharp knife and tweezers. The piece of the cuticle was mounted on a glass slide with double-sided tape for the Mueller-matrix spectroscopic ellipsometry measurements which were performed with a dual rotating compensator ellipsometer (J. A. Woollam Co., Inc.). Since the cuticle is curved, focusing probes were used to achieve a beam spot with size below 100 μm. More details about the instrument can be found in references [18-21]. The measurements were performed at angles of incidence between 25 and 75° in steps of 5° in the wavelength range of 245 to 1000 nm. Scanning electron microscopy (SEM) images of the cuticle cross-section were acquired with a LEO 1550 Gemini system. The samples were mounted on double-side copper tape and coated with a thin platinum layer of around 2 nm to obtain a conductive surface.

The most general description of the polarization and depolarization capability of light induced by reflection from a sample, is given by its 4×4 reflection Mueller matrix,
\[
M = \begin{bmatrix}
    m_{11} & m_{12} & m_{13} & m_{14} \\
    m_{21} & m_{22} & m_{23} & m_{24} \\
    m_{31} & m_{32} & m_{33} & m_{34} \\
    m_{41} & m_{42} & m_{43} & m_{44}
\end{bmatrix},
\]

which relates the Stokes vectors of the incident (\(S_i\)) and reflected (\(S_r\)) beams according to the relationship \(S_r = MS_i\). The Stokes vectors have the components,

\[
S = \begin{bmatrix}
    I_p + I_s \\
    I_p - I_s \\
    I_{+45} - I_{-45} \\
    I_R - I_L
\end{bmatrix},
\]

where \(I_p, I_s, I_{+45},\) and \(I_{-45}\) are, respectively, the irradiances of polarized light components parallel (p), perpendicular (s), at +45° and at -45° with respect to the plane of incidence; \(I_R\) and \(I_L\) are the irradiances of right- and left-handed circularly polarized light components. In this work, normalized Mueller matrices \((m_{11}=1)\) and \(S_i\) \((I_p+I_s=1)\) are used.

In the particular case of unpolarized incident light \(S_i = [1,0,0,0]^T\) the reflected light beam is given by \(S_r = [1,m_{21},m_{31},m_{41}]^T = [1,P]^T\), where \(P=[m_{21},m_{31},m_{41}]^T\) is the so-called polarization vector of \(M\) and \(T\) indicates transpose. Since the reflected light in general will be partially polarized, the degree of polarization \(P\) for incident unpolarized light can be calculated from [18],

\[
P = \sqrt{m_{21}^2 + m_{31}^2 + m_{41}^2}.
\]

In general, the polarized part of the reflected irradiance will be elliptically polarized with ellipticity \(e=\frac{b}{a}\) where \(a\) and \(b\) are the major and minor axes of the polarization ellipse, respectively. The major axis is located at an azimuth angle \(\phi\) measured from the plane of incidence. The parameters \(e\) and \(\phi\) are given by [18],

\[
e = \tan \left( \frac{1}{2} \arcsin \left( \frac{m_{41}}{\sqrt{m_{21}^2 + m_{31}^2 + m_{41}^2}} \right) \right),
\]

and

\[
\phi = \frac{1}{2} \arctan \left( \frac{m_{31}}{m_{21}} \right).
\]

On the other hand, the average measure of the depolarization produced by a system for all incident pure states is given by the depolarizance [24],

\[
D = 1 - \left[ \frac{1}{3} \left( \frac{\text{tr}(M^T M)}{m_{11}^2} - 1 \right) \right]^{1/2}.
\]

where in our case \(m_{11}=1\).

3. Results and discussion

3.1 Cuticle microstructure

The cuticle of the beetle is comprised of the epicuticle, the exocuticle and the endocuticle. Figure 1 shows a SEM image of a cross section of the beetle cuticle revealing the epicuticle
and exocuticle comprising a layer around 15 μm thick. The thickness of the epicuticle (Ep) is about 100 nm for the specimen under study. Two regions can be differentiated in the exocuticle: the outer exocuticle (o-Ex) with a fine multilayer structure and the inner exocuticle (i-Ex) also with a multilayer structure. The microstructure observed in Fig. 1 is representative of other species where the selective Bragg reflection is located at the outer exocuticle [1]. In some of them, the inner exocuticle is tanned with a black/brown pigment as in C. mutabilis. Beneath the inner exocuticle is the endocuticle which provides mechanical support.

![Green specimen of C. mutabilis and SEM image of a cross section of its cuticle on the abdominal side. The epicuticle (Ep) as well as the outer (o-Ex) and inner (i-Ex) parts of the exocuticle are shown.](image)

Fig. 1. Green specimen of C. mutabilis and SEM image of a cross section of its cuticle on the abdominal side. The epicuticle (Ep) as well as the outer (o-Ex) and inner (i-Ex) parts of the exocuticle are shown.

### 3.2 Polarization properties of the cuticle of C. mutabilis

The features of the Mueller matrix (not shown) of the green specimen studied in this work is similar to that previously reported for a red specimen, i.e. it exhibits the symmetry properties of chiral systems [21]. Here, we first focus on the polarization properties \((P, e, \varphi)\) of light reflected from the beetle cuticle under unpolarized incident light calculated from Eqs. (3)-(5) and the depolarizance of the experimental Mueller matrix from Eq. (6). These four quantities are shown in Fig. 2 in contour polar representations where the radial and angular coordinates correspond to the wavelength \(\lambda\) and angle of incidence \(\theta\), respectively. Noteworthy in Fig. 2 is the behavior of \(P, e, \varphi,\) and \(D\) in a narrow spectral range which corresponds to the band of selective Bragg reflection. Referring to Figs. 2(a) and 2(b), we observe that at small angles of incidence \((\theta<40^\circ)\) and in the spectral range of 500 to 600 nm, unpolarized incident light is reflected with a relatively high degree of polarization \((P>0.8)\), left-handed polarized and with elliptical to near-circular character \((e<0.5)\). In Fig. 2(c), \(|\varphi|\) shows a strong variation with wavelength and angle of incidence. It can be noticed that the Bragg reflection band shifts to shorter wavelengths as \(\theta\) increases. Outside the spectral range of Bragg reflection, \(e=0\) and \(|\varphi|=90^\circ\) for all angles of incidence. Since in addition, the degree of polarization \(P\) is high for \(45^\circ<\theta<70^\circ\), linear s-polarization is dominating the light reflected at these conditions. Indeed, a maximum in \(P=1\) is noticed around \(\theta=55^\circ\) which presumably indicates a pseudo-Brewster’s angle. Strictly, at the Brewster angle \(e=0, |\varphi|=90^\circ\) and \(P=1\).

The depolarizance of the measured Mueller matrix is shown in Fig. 2(d). According to its definition, \(D\) is interpreted as unity minus the polarimetric purity of \(M\) [24]. Thus, for an ideal depolarizer \(D=1\) whereas the minimum value \(D=0\) corresponds to a non-depolarizing system and \(M\) is termed a Mueller-Jones matrix. In the spectral range of Bragg-like reflection in Fig. 2(d), \(D\) is a rapidly varying function of wavelength showing sharp maxima but, as can be
noticed, on the average $D\approx0.2$ and $\mathbf{M}$ moderately deviates from a non-depolarizing system. Nevertheless, in the complementary spectral range $D<0.05$ and $\mathbf{M}$ represents a nearly non-depolarizing system. Non-uniformity in cuticle thickness in the measured area, surface and volume irregularities, and lateral inhomogeneities in the cuticle are examples of possible causes of light depolarization.

Fig. 2. Properties of polarized light reflected from a green C. mutabilis for unpolarized incident light: (a) degree of polarization, (b) ellipticity, and (c) azimuth. The depolarizance of the Mueller matrix is shown in (d).

3.3 Dispersion relation and Bragg-like reflection

In the Mueller matrix spectra of C. mutabilis it is noticeable that near and at the spectral range of selective reflection, the elements of $\mathbf{M}$ present strong interference oscillations as was previously reported for a red specimen [21]. For the green specimen studied in this work, such characteristic is exemplified in Fig. 3 with the elements of the polarizance vector at three angles of incidence. For the analysis, the dependence on photon energy is used here. The presence of these interference oscillations is indicative of highly transparent materials comprising the beetle cuticle. Considering the spectra of $m_{33}$ and $m_{11}$ at $\theta=25^\circ$ in Fig. 3, four spectral regions can be distinguished from low to high photon energies: I) high frequency interference oscillations, II) slowdown of frequency, III) oscillations of low frequency with decreasing amplitude, and IV) quenching of interference oscillations. At $\theta=25$ and $50^\circ$, features in $m_{33}$ characterize the three boundaries separating these four spectral regions: I-II onset of selective reflection of left-handed polarized light ($m_{33}<0$); II-III a peak in $m_{33}$ appears; III-IV cut-off of the selective reflection band. It can be noticed that increasing the angle of incidence the amplitude of the interference oscillations decreases (mostly in region I) and the boundaries shift to higher photon energies. At large angles of incidence and for incident unpolarized light, right-handed polarized light ($m_{33}>0$) is reflected from the beetle cuticle in a narrow spectral range as shown for $\theta=75^\circ$. Previously, it was reported that $m_{33}>0$ in the beetle Chrysina argenteola at $\theta>45^\circ$ [18].
In order to extract quantitative information from the interference patterns in the elements of $\mathbf{M}$, we consider the procedure commonly used in optical analysis of isotropic thin films. For incident light at an angle $\theta$ from an ambient with refractive index $n_i$, multiple reflections inside a non-absorbing film of thickness $d$ and refractive index $n_\omega$, produce maxima or minima in the optical measurements when the phase factor $\beta$ equals a multiple integer of $\pi$ [25], that is:

$$\beta = 2K|| d = \frac{4\pi d}{\lambda} \sqrt{n_\omega^2 - n_i^2} \sin^2 \theta = m\pi,$$

where $m$ is an integer and $K||$ is the component of the wave vector parallel to the $z$-axis, which is perpendicular to the sample surface ($xy$-plane). Since $K||$ depends on the photon energy $E$, we first investigate the corresponding dependence for $m$. However, as the actual values of $m$ are unknown, we introduce a temporary index $m$. Thus, starting from the low photon energy side, the first clear maximum or minimum in $m_{31}$ is located and arbitrary labeled $m=1$. Subsequent maxima and minima are consecutively indexed with integer values of $m$. Figure 4(a) shows the dependence of $m$ on photon energy for several angles of incidence. It is noteworthy that all data sets shown in Fig. 4(a) exhibit three clear regions; each showing a near-linear dependence with photon energy but with different slope. The first discontinuity in slope corresponds to the change in the frequency of the interference oscillations at boundary I-II already noticed in Fig. 3.

The arbitrariness in the choice of the first maximum or minimum, that is, corresponding to $m = 1$ can be removed by an offset correction. For that, the apparent linear behavior of the data with $E$ in Fig. 4(a) at low photon energies was fitted with a linear relationship $m(E) = c_1 + c_2 E$ obtaining the two constants $c_1$ and $c_2$. Thus, the offset correction shown in Fig. 4(b) implies $m = m_0 - m_0$, where $m_0$ is the closest integer to $c_1$. For clarity, only data for the smallest and largest angles of incidence are shown in Fig. 4(b); the dashed lines correspond to extrapolation to zero photon energy using $m(E) = c_2 E$. According to Eq. (7), the results shown in Fig. 4(b) provide evidence for the dispersion relation of $K||$ in the cuticle of the beetle.
C. mutabilis. The data in Fig. 4(c) are the results obtained by applying the same procedure to data from a red specimen whose Mueller matrix was previously analyzed [21]. As is shown in the next section, the data in Figs. 4(b) and 4(c) resemble the dispersion relation of optical modes found in ChNLC structures. Some structural parameters of the beetle cuticle can now be estimated. First, by using the data at low photon energies in Fig. 4(b), the thickness of the region in the cuticle producing the interference oscillations can be estimated from Eq. (7) where \( n_{av} \) is interpreted as the effective refractive index of the beetle cuticle at long wavelengths. From the data at \( \theta=25^\circ \), with air as ambient \( (n=1) \) and for \( n_{av}=1.54 \) [19], the estimated thickness is \( d=10.3 \) \( \mu \)m in agreement with the thickness of the outer exocuticle determined from the SEM image in Fig. 1. Furthermore, in Fig. 3 the band where \( m_{av}<0 \) is centered around \( \lambda_0=560 \) nm which can be used to estimate the pitch \( \Lambda \) to 394 nm according to \( \Lambda = \lambda_0 \cos \theta \) [11]. Thus, the number of periods in the outer exocuticle is \( d/\Lambda=26.1 \). For the red specimen the estimated thickness was found to be 7.5 \( \mu \)m [21]. This difference in thickness explains the different slopes observed in Figs. 4(b) and 4(c).

![Dispersive Relation](image)

**Fig. 4.** (a) Spectral position of maxima and minima in \( n_{av} \) of a green specimen for \( \theta \) between 25\(^\circ\) and 75\(^\circ\) in steps of 10\(^\circ\) from left to right. Dispersion relations in Eq. (7) for \( \theta=25^\circ \) and 75\(^\circ\) after offset correction for (b) the green specimen studied in this work and (c) the red specimen previously studied [21].

### 3.4 Dispersion relation of optical modes in ChNLC structures

In this section we provide support that evidences the data in Fig. 4(b) as a dispersion relation. For that, we adopt the framework developed for the propagation and polarization states of optical modes in ChNLC established some decades ago [15-17]. Briefly, the ChNLC is considered to be comprised of anisotropic non-absorbing layers with a local dielectric tensor diag[\( \varepsilon_1, \varepsilon_2, \varepsilon_3 \)], as illustrated in Fig. 5(a). Alternatively, the principal refractive indices \( n_j=(\varepsilon_j)^{1/2} \) \((j=1,2,3)\) can be used. The continuous rotation of the principal axes \( \varepsilon_1 \) and \( \varepsilon_2 \) defines the pitch \( \Lambda \) after completing 360\(^\circ\), as is schematically shown in Fig. 5(b) for \( \varepsilon_1 \). Let us consider the oblique incidence of an electromagnetic wave of wavevector \( K_\parallel \) to the ChNLC from a medium of refractive index \( \eta_1 \) as shown in Fig. 5(b). The plane of incidence coincides with the \( xz \)-plane. Inside the ChNLC structure, the wave vector \( K \) has components parallel \( (K_\parallel) \) and perpendicular \( (K_\perp) \) to the axis of the helix \( (z\text{-axis}) \). The former is normalized according to \( k = K_\parallel (4\pi/\Lambda) \) whereas the latter is given by \( K_\perp (2\pi/\lambda) = \eta \sin \theta \). The periodicity \( (\Lambda/2) \) along the helix allows looking for solutions represented as an infinite sum of plane waves having wave vectors \( K_j = K + 2l/q \), where \( l \) is an integer and \( q \) is a reciprocal lattice vector parallel to the helix of magnitude \( q=2\pi/\Lambda \). The substitution of that representation in Maxwell equations leads to certain recurrent relations for the amplitudes of the plane waves. Thus, a set of homogeneous equations is obtained and the roots of the characteristic equation determine
the solutions for $k$ (and hence for $K_{||}$). Only two solutions ($k_{1,2}$) are independent because waves propagating in the forward ($k^+$) and backward ($k^-$) directions are simply related by $k_{1,2}^\pm = \pm k_{1,2} + l$. Three general types of solutions are found: A) The wave vector $k$ is real-valued and the waves propagate without attenuation; B) The wave vector is complex $k=k'+ik''$ indicating damped waves with attenuation length $\eta=\Lambda/(4\pi k'')$ and $k'$ equaling a half integer $(n/2)$ which expresses the condition for selective Bragg reflection of order $n$ ($K_{||}=2\pi n/\Lambda$); C) Two roots of the characteristic equation are conjugate each other and the real part does not satisfy the Bragg condition. Particularly, in the long-wave limit the solutions are real-valued and can be expressed as [15-17],

\[
k_a = \frac{\Lambda}{2\lambda} \sqrt{\varepsilon_m \varepsilon_3 - n^2 \sin^2 \theta} \quad k_b = \frac{\Lambda}{2\lambda} \sqrt{\varepsilon_m - n^2 \sin^2 \theta}
\]

where $\varepsilon_m=(\varepsilon_1+\varepsilon_2)/2$ is the in-plane average dielectric constant. The wave vectors $k_a$ and $k_b$ in Eq. (8) can be identified, respectively, with those of linearly p- and s-polarized waves, i.e. they correspond to wave propagation in an uniaxial medium with ordinary $\varepsilon_m$ and extraordinary $\varepsilon_3$ components [25].

![Diagram](attachment:image.png)

**Fig. 5.** (a) Anisotropic layers with local principal components $\varepsilon_1$, $\varepsilon_2$, and $\varepsilon_3$ comprising the CHNCL structure; the counter-clockwise rotation of $\varepsilon_3$ (and $\varepsilon_2$) around the axis of the helix (z) generates a left-handed chiral structure. (b) Wave vectors geometry for oblique incidence on a CHNLC structure; the full rotation of 360° of the principal axes defines the pitch $\Lambda$.

The properties of the first-order reflection band can be determined within a two-wave approximation through the solutions of the characteristic equation [17],

\[
\left(x^2 - x_a^2\right)^2 - x_b^2 \left(x^2 - x_a^2\right) - x_c^2 \left(x^2 - x_a^2\right) = 0,
\]

where

\[
x = k - 1/2, \quad x_{a,b,c} = k_{a,b,c} - 1/2, \quad X_{a,b,c} = k_{a,b,c} + 1/2
\]
or oblique incidence are in ice (a),
or a given

Figure 6(a) shows the dispersion relation of the normalized complex wave vector \( k = k' + i k'' \) in a ChNLC structure calculated from Eq. (9) for \( \theta = 25 \) and \( 75^\circ \). The incident medium is air \((n=1)\) and the ChNLC parameters are \( \Lambda = 390 \text{ nm}, \ n_1=1.58 \) and \( n_1=n_3=1.5 \), typical values found in beetle cuticle [19]. For simplicity, the dispersion in the refractive indexes was not considered. As can be observed in Fig. 6(a), in most parts of the spectral range the solutions are real-valued indicating modes propagating without attenuation (type A). At low photon energies, the two real-valued solutions \( k_1 \rightarrow k_a \) and \( k_2 \rightarrow k_b \), i.e. linearly p- and s- polarized waves, respectively, are obtained, whereas at higher photon energies the reverse situation holds \( k_1 \rightarrow k_b \) and \( k_2 \rightarrow k_a \). For each angle of incidence there is a narrow band where at least one of the solutions \( (k_2) \) is complex and the Bragg condition \( k_2' = 1/2 \) is satisfied (type B). In other words, the effective wavelength \( \lambda_{\text{eff}} \) inside the ChNLC structure associated to \( k_1 \) is such that \( \lambda_{\text{eff}} = \Lambda \). Regarding the imaginary part of the wave vector, we see in Fig. 6(a) that \( k_2'' \) peaks at 2.15 and 2.67 eV for \( \theta = 25 \) and \( 75^\circ \), respectively. For \( \theta = 75^\circ \) both \( k_1'' \) and \( k_2'' \) present a maximum located approximately at the same photon energy.

Fig. 6(b) shows a magnification of \( k' \) for \( \theta = 25, 50 \) and \( 75^\circ \). It can be observed that as \( \theta \) increases the first-order characteristic reflection band widens and shifts toward higher photon energies [14,15]. Other characteristics of the reflection band largely depend on the refractive indices. For simplicity, we restrict the discussion to uniaxial systems and for a given \( \theta \) the anisotropy parameter \( \Delta n = n_1 - n_2 \) determines the spectral width of the reflection band [9,13]. Solutions of type C for the wavevector appear at large angles of incidence when locally biaxial slabs are considered as those deduced from regression analysis of Mueller-matrix data of Cetonia aurata [19]. If normal dispersion in the refractive indices is considered, the reflection bands at large angles of incidence shift to lower photon energies. Instead of the imaginary part \( n_{\text{eff}} \) of the wavevector \( k_2 \), the attenuation length is shown in Fig. 6(c) which clearly diverge at band edges. The minimum attenuation lengths for \( \theta = 25, 50 \), and \( 75^\circ \) are \( n_{\text{min}} = 2.3, 2.1, \) and 1.94 \( \text{nm} \), respectively. Larger values of \( \Delta n \) produce smaller values of \( n_{\text{min}} \).

The polarization states of light waves in ChNCL structures for oblique incidence are in general elliptic [16]. The nature of eigenmodes ranges from near circular (left- and right-handed) at small angles of incidence, to near linear (p- and s- polarized) at medium and large angles. The latter result is in agreement with the polarization properties of the light reflected from the beetle cuticle for unpolarized incident light as was shown in panels (a)-(c) of Fig. 2; a decreased ellipticity of left-handed polarized light in the reflection band (Fig. 2(b)) with increase of \( \theta \) and a high degree of linear s-polarization for \( 45^\circ < \theta < 70^\circ \) and wavelengths outside the Bragg-reflection band. Since at the latter conditions the cross-polarized reflectance \( R_{pr} \) has been shown to be near zero [21], unpolarized incident light excites the mode \( k_2 (k_1) \) at photon energies lower (higher) than the Bragg-reflection band.

and

\[
a' = \frac{1}{2} \frac{2 \epsilon_3 - n_i^2 \sin^2 \theta}{\sqrt{\epsilon_3 - n_i^2 \sin^2 \theta}}, \\
c = \frac{(\epsilon_1 - \epsilon_3) k_0^2}{8 k_0^2 \sqrt{\epsilon_3 - n_i^2 \sin^2 \theta}}, \\
k_c = \sqrt{\frac{k_a^2 + k_p^2 \epsilon_3 - k_b^2 n_i^2 \sin^2 \theta}{2 \epsilon_3 - n_i^2 \sin^2 \theta}}.
\]
3.5 Structural model for the exocuticle of *C. mutabilis*

The broadening of the selective reflection band of ChNLC has been achieved by using structures of multiple pitches (discrete or graded) inspired by ideas from nature [9]. Indeed, structures with a jump in the pitch have been imaged for beetle cuticles [4,5]. Following this idea, let us consider the helical structure shown in Fig. 7(a) which is comprised of slabs organized in two helical stacks of pitches \( \Lambda_1 \) and \( \Lambda_2 \) and thicknesses \( d_1 \) and \( d_2 \). The outer exocuticle of *C. mutabilis* (thickness \( d_1+d_2 \)) is represented by these two helical stacks. Since the same composition is expected through the whole outer exocuticle, the same local dielectric tensor is assumed for slabs in these two chiral stacks. The substrate in Fig. 7(a) represents the (sclerotized) inner exocuticle which in *C. mutabilis* is tanned with a black-brown pigment.

In order to discuss the propagation of light in the stacked ChNLC-like structures of Fig. 7(a), the dispersion relation of optical modes for each semi-infinite structure was calculated. The pitches and refractive indexes considered were \( \Lambda_1=390 \) nm, \( \Lambda_2=360 \) nm, \( n_1=1.58 \), and \( n_2=n_1=1.5 \). For clarity, in Figs. 7(b) and 7(c) only the dispersion relation for the real part of the optical mode with wave vector satisfying the Bragg condition \( k_2 \) is shown for \( \theta=25^\circ \) and 75°. The corresponding attenuation lengths \( \eta_1 \) and \( \eta_2 \) are shown in Figs. 7(d) and 7(e). As
expected, a smaller pitch shifts the first-order reflection band to higher photon energies. Four spectral regions can be identified in correspondence with those in Fig. 3.

In the spectral region I, \( k_2 \) is real-valued in both chiral stacks and electromagnetic waves of effective wavelength \( \lambda_{\text{eff}} > \Lambda_{1,2} \) propagate without attenuation throughout the whole helical double-structure. In this case, the total phase change of light reflected is given by,

\[
\beta_1 = 2K_{||}d_{1} + 2K_{\perp}d_{2}. \tag{11}
\]

Since the total thickness of the outer exocuticle is probed by the light beam, high frequency oscillations are expected in the reflected spectra regardless of the incidence angle. For photon energies in region II, the Bragg condition is met in stack 1 and left-handed polarized light is reflected. For \( d_{1} \) sufficiently larger than \( \eta_{1} \), the structure below stack 1 is not probed by the light beam. In the spectral region III, waves propagate without attenuation in stack 1 (probing the whole thickness \( d_{1} \)) but selective reflection is observed from stack 2 broadening the band where \( m_{11} > 0 \) as shown in Fig. 3 for \( \theta=25^\circ \). Since in stack 2 the light beam probes the attenuation length \( \eta_{2} \), the phase change in this case is,

\[
\beta_{\text{in}} = 2K_{||}d_{1} + 2K_{\perp}d_{2}. \tag{12}
\]

If \( d_{2} > \eta_{2} \), the depth probed by the light beam is smaller than the total thickness of the outer exocuticle. Therefore, the frequency of interference oscillations in region III is lower than in region I. This latter result explains the difference in slopes noticed in the experimental data shown in Figs. 4(b) and 4(c). The peak seen in \( m_{11} \) of Fig. 3 for \( \theta=25^\circ \) and marked with the arrow at the boundary between regions II and III, can be explained as defect mode since \( K_{||} \) is real-valued in both chiral stacks and the light waves propagate without attenuation in a very narrow spectral range. The case of region IV is similar to the situation found for region I where \( K_{||} \) is real-valued in both stacks. However, in region IV \( \lambda_{\text{eff}} < \Lambda_{1,2} \) and irregularities in beetle cuticle affect the coherency of light waves which smears out the interference oscillations. In spite of the overlap of region IV at \( \theta=25^\circ \) and region I at \( \theta=75^\circ \), at grazing incidence the interference oscillations are observed because at this condition \( \lambda_{\text{eff}} > \Lambda_{1,2} \). Similar arguments can be applied for a model with more chiral stacks in the structure.

![Fig. 7. (a) Schematics of wave propagation in a ChNLC-like structure with two chiral stacks of pitches \( \Lambda_{1} \) and \( \Lambda_{2} \) of of thicknesses \( d_{1} \) and \( d_{2} \). (b) and (c) Real part of the wave vector of the optical mode producing Bragg-like reflection at angles of incidence 25° and 75°, respectively; (d) and (e) show the corresponding attenuation lengths. The dispersion relations were calculated for semi-infinite uniaxial structures with pitches \( \Lambda=390 \) and 360 nm and refractive indices \( n_{1}=1.58 \) and \( n_{2}=n_{1}=1.5 \).](image-url)
Finally, in Fig. 3 it was noted that $m_{41}>0$ at large angles of incidence in region III. Two possibilities are suggested to explain the reflection of right-handed polarized light with ellipticity $e>0$ from a left-handed structure. The first explanation is similar to that given for reflection of right-handed polarized light in Plusiotis (now Chrysina) resplendens [26]. As was mentioned above, at large $\theta$ the polarization of the optical modes acquires a more linear character and stack 1 might behave as a half-wave plate for some wavelengths. Thus, during the propagation through stack 1 incident right-handed polarized light could be transformed into the left-handed which would be reflected from stack 2. As it travels back through stack 1 the emerging light would be observed as right-handed. The effectiveness of stack 1 as a half-wave plate would depend on the relative orientation of the plane of incidence with respect to the segments comprising the abdomen of C. mutabilis (Fig. 1) because the strength of positive values of $m_{41}$ showed that dependence [21]. The other possibility is related to the incidence of light from a low refractive index medium (air) to beetle cuticle. In liquid crystals cells the chiral nematic phase is sandwiched between symmetric media (glass) typically with a refractive index similar to the average of the liquid crystal. Nevertheless, calculations have shown that changing the external media from glass to air, $e$ shows opposite signs below and above the band of selective reflection at large angles of incidence [27].

4. Conclusions

The polarization properties of the cuticle of the scarab beetle C. mutabilis and allowed and forbidden optical modes in the cuticle have been studied using Mueller matrix spectroscopic ellipsometry. It was shown that for unpolarized incident light, the reflected beam is left-handed polarized with a high degree of circular polarization in a narrow spectral band at small angles of incidence. At intermediate angles of incidence and outside of the Bragg reflection band, the reflected beam has a strong linear s-polarized character. Clear interference oscillations in the spectra are due to the coherent superposition of reflected light from the cuticle interfaces. Structural parameters for the outer exocuticle were estimated from features in the spectra: a thickness of 10.3 $\mu$m, a pitch of 390 nm and 26 periods in the helix were found. It was also found that the spectral position of maxima and minima of interference oscillations gives insight into the dispersion relation of the optical modes in the beetle cuticle. A dispersion relation, calculated within a two-wave approximation originally developed for ChNCL structures, supports the experimental evidence. A structural model comprised of two chiral stacks of uniaxial slabs was derived from a theoretical analysis of wave propagation in the beetle cuticle.

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