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Theoretical reflectances and phase shifts at achiral-chiral interfaces

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Theoretical reflectances and phase shifts at achiral-chiral interfaces. The complex Fresnel coefficients corresponding to the reflection of plane waves at planar achiral-chiral interfaces have been calculated as functions of angle of incidence, in order to show the noticeable features that could be exploited in instrumental or operational reflection techniques.

Theoretische Reflexionskoeffizienten und Phasenschiebungen an achiral-chiralen Grenzflächen. Um die bemerkenswerten Vorteile zu zeigen, die bei der Auswertung von instrumentellen oder operationellen Reflexionstechniken möglich sind, werden die komplexen Fresnelkoeffizienten bei der Reflektion von Planwellen an planaren achiral-achiralen Grenzflächen als Funktion des Einfallswinkels berechnet.

1. Introduction

A variety of methods used to ivestigate optical properties of materials are based on measuring changes in the state of polarization of light by reflection or transmission [1]. A change of polarization takes place on reflection at an interface due to the difference in amplitude attenuation or phase shift experienced by two mutually orthogonal polarization components. In order to interpret ellipsometric data it is necessary to know the behavior of the complex amplitude reflection coefficients in terms of the macroscopic optical properties that characterize the media at each side of the interface. Usually one medium (the ambient) is of known optical properties and the other (the substratum) is a bulk phase of the material under investigation. A considerable amount of theoretical and experimental work has been done on this topic for achiral-achiral boundaries, particularly when light is incident from a transparent dielectric onto the surface of another transparent dielectric, a semiconductor or a metal [1, 2].

A chiral medium [3] is characterized by either a left- or a right-handedness in its microstructure. Left- and right-circularly polarized fields propagate with different phase velocities and so the vibration ellipse of light is transformed upon passage through such a medium. This phenomenon of natural optical activity has attracted a great amount of attention since it is exhibited by a multitude of organic molecules at optical frequencies.

Every noticeable feature in the curves representing the variation of the reflectivity and phase shifts with angle of incidence, when there is no chirality involved, has been exploited in instrumental or operational *reflection* ellipsometric techniques. The manifestations of chiral asymmetries, on the other hand, have been investigated almost exclusively using light *transmission* method as, for example, in the measurement of optical rotatory dispersion or circular dichroism [4]. But transmission methods are not always adequate: if the transmitted beam is not experimentally acessible or the sample is not transparent, reflection methods must be used. In order to develop reflection ellipsometric techniques when chirality is involved, it would be desirable to exhibit the complete behavior of the complex reflection coefficients, a matter that we will face here.

Theoretical and experimental work on the reflection problem at planar achiral-chiral interfaces can be found in [5–10]. Fresnel coefficients were obtained in connection with the controversy about the appropriate set of constitutive relations characterizing optically active media [7] or in the discussion of the Brewster law for achiral-chiral interfaces [10]. Curves showing the relative intensity of reflected light as a function of incident angle and for different polarizations have been calculated [7, 8], but not curves for each element of the reflection matrix. Furthermore, only intensities or magnitudes and not the phases of the fields have been analyzed.

The phase of the reflected fields could have great practical importance in the ellipsometric or interferometric characterization of chiral materials. To study the theoretical behavior of the complex amplitudes of the fields reflected at an achiral-chiral interface we first give the corresponding reflection matrix and then we obtain both magnitude and phase curves as functions of angle of incidence and polarization for co- and cross-polarized reflected waves. Not only it is possible to observe in the phase curves the same features as in the achiral case, but also new ones related to the fact that the two transmitted modes in the chiral medium become evanescent at different angles of incidence. The evolution of the remarkable points in the curves when the chirality parameter is varied is also studied.

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2. Reflection matrix

Let the $x - y$ plane be the interface between a nongyrotropic medium (region $z < 0$, permittivity ε_0 and permeability μ_0) and an optically active medium (region $z > 0$) characterized by the Drude-Born-Fedorov [3] constitutive relations:

$$\begin{aligned} \boldsymbol{D} &= \varepsilon (\boldsymbol{E} + \beta \nabla \times \boldsymbol{E}) \\ \boldsymbol{B} &= \mu (\boldsymbol{H} + \beta \nabla \times \boldsymbol{H}). \end{aligned} \tag{1}$$

We choose a representation of incident and reflected waves in terms of linearly polarized components having their electric field parallel (P) or perpendicular (S) to the plane of incidence. A plane wave of arbitrary polarization in incident from the $z < 0$ region at angle ϑ_0 with the z-axis

$$\boldsymbol{E}_i = [A_i^s \hat{y} + A_i^p (-\hat{x} \cos \vartheta_0 + \hat{z} \sin \vartheta_o)] \, e^{i(\varkappa x + \alpha_0 z)} \tag{2}$$

where $\varkappa = k_0 \sin \vartheta_0$, $k_0 = \omega \sqrt{\mu_0 \varepsilon_0}$, $\alpha_0 = \sqrt{k_0^2 - \varkappa^2} = k_0 \cos \vartheta_0$ and harmonic time dependence $\exp(-i\omega t)$ has been used. It produces a reflected wave

$$\boldsymbol{E}_r = [A_r^s \hat{y} + A_r^p (\hat{x} \cos \vartheta_0 + \hat{z} \sin \vartheta_o)] \, e^{i(\varkappa x - \alpha_0 z)} \tag{3}$$

and a transmitted wave which has to be expressed in terms of the propagation modes in the chiral medium, i.e. left (subscript 1) and right (subscript 2) circularly polarized waves*:

$$\begin{aligned} \boldsymbol{E}_t = {} & B_1^+ \left[\hat{y} + i \left(-\hat{x} \frac{\alpha_1}{\gamma_1} + \hat{z} \frac{\varkappa}{\gamma_1} \right) \right] e^{i(\varkappa x + \alpha_1 z)} \\ & + B_2^+ \left[\hat{y} + i \left(\hat{x} \frac{\alpha_2}{\gamma_2} + \hat{z} \frac{\varkappa}{\gamma_2} \right) \right] e^{i(\varkappa x + \alpha_2 z)} \end{aligned} \tag{4}$$

In this representation γ_1 and γ_2 are the mode wavenumbers

$$\gamma_1 = \frac{k}{1 - f} \tag{5}$$

$$\gamma_2 = \frac{k}{1 + f} \tag{6}$$

and we have defined $k = \omega \sqrt{\mu \varepsilon}$ and $f = k \beta$. The other symbols appearing in (4) are

$$\alpha_1 = \sqrt{\gamma_1^2 - \varkappa^2} = \gamma_1 \cos \vartheta_1 \tag{7}$$

$$\alpha_2 = \sqrt{\gamma_2^2 - \varkappa^2} = \gamma_2 \cos \vartheta_2 . \tag{8}$$

Phase matching at $z = 0$ gives the angles ϑ_1 and ϑ_2 that each transmitted wavevector makes with the z-axis

$$\varkappa = k_0 \sin \vartheta_0 = \gamma_1 \sin \vartheta_1 = \gamma_2 \sin \vartheta_2 . \tag{9}$$

Imposition of Maxwell boundary conditions at the interface leads to a linear relation between the reflected and incident field amplitudes which can be written in matrix notation as [10]

$$\begin{bmatrix} A_r^s \\ A_r^p \end{bmatrix} = \begin{bmatrix} R_{ss} & R_{sp} \\ R_{ps} & R_{pp} \end{bmatrix} \begin{bmatrix} A_i^s \\ A_i^p \end{bmatrix} . \tag{10}$$

The matrix coefficients are:

$$R_{ss} = \frac{2}{\Delta} [2\xi (\delta_1 \delta_2 - 1) + (\xi^2 - 1)(\delta_1 + \delta_2)] \tag{11}$$

$$R_{pp} = \frac{2}{\Delta} [2\xi (\delta_1 \delta_2 - 1) - (\xi^2 - 1)(\delta_1 + \delta_2)] \tag{12}$$

$$R_{sp} = -R_{ps} = -4i \frac{\xi}{\Delta} (\delta_1 - \delta_2) \tag{13}$$

with

$$\delta_1 = \frac{(\alpha_1 k_0)}{(\alpha_0 \gamma_1)}, \qquad \delta_2 = \frac{(\alpha_2 k_0)}{(\alpha_0 \gamma_2)}, \qquad \gamma = \left[\frac{\mu_0 \varepsilon}{\mu \varepsilon_0} \right]^{1/2} \quad \text{and}$$

$$\Delta = (\xi - 1)^2 (\delta_1 - 1)(\delta_2 - 1) - (\xi + 1)^2 (\delta_1 + 1)(\delta_2 + 1) . \tag{14}$$

It should be noted that β having positive real part corresponds to a right handed medium. The replacement of β by $-\beta$ in the above equations exchanges the roles of subscripts 1 and 2, leaving the copolarized reflected components unchanged but changing the sign of the cross-polarized components. So the determination by reflection of the handedness of the medium (i.e., the sign of β) necessarily involves the measurement of the phase of the cross polarized component.

In the following we will assume that the chiral medium is right handed so that $\gamma_1 > \gamma_2$ while β is positive real. The geometric description of the transmitted waves is obtained from eq. (9).

3. Results

From the geometric and energetic point of view it is convenient to distinguish three cases which correspond to three different behaviors.

Case 1)

There are always real angles ϑ_1 and ϑ_2 of refraction for every angle of incidence ϑ_0, if the constitutive parameters satisfy the following condition

$$\sqrt{\mu_0 \varepsilon_0} < \frac{\sqrt{\mu \varepsilon}}{1 + f} . \tag{15}$$

The curves of R_{pp} and R_{ss} as functions of incidence angle (not shown) are very similar to the ones obtained in the dielectric-dielectric case. $|R_{pp}|$ and $|R_{ss}|$ coincide at normal and at grazing incidence; $|R_{ss}|$ increases monotonically and $|R_{pp}|$ first decreases from normal incidence up to an angle ϑ_0 where it becomes zero and then increases from zero to one (normal incidence). Since α_1 and α_2 are real numbers, there are no interesting features in the phase curves when condition (15) is satisfied. The phases of R_{ss} and R_{sp} are π and $\pi/2$ respectively for all angles of incidence and the phase of R_{pp} is zero or π depending on

* This definition of handedness of waves is the classical one, and is the reverse of the IEEE definition.

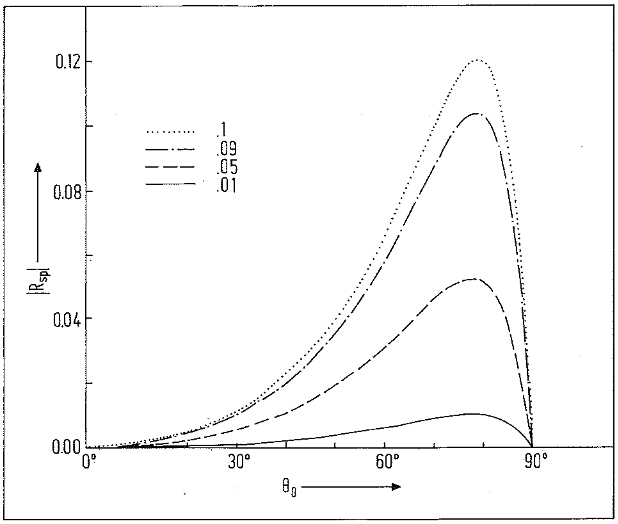

Fig. 1. Plots of $|R_{sp}|$ for $\mu = \mu_0$, $\varepsilon = 1.33\,\varepsilon_0$ and $f = k\beta$ in the range [0, 0.1] versus angle of incidence θ_0.

whether ϑ_0 is smaller or greater than $\tilde{\vartheta}_0$. We also observed that $\tilde{\vartheta}_0$ is not much sensitive to variations of the chirality parameter f in the range [0, 0.1]. As expected, the cross polarization terms are the most sensitive to chirality. In fig. 1 we show the curves for $|R_{sp}|$ when $\mu = \mu_0$, $\varepsilon = 1.33\,\varepsilon_0$ and for different values of f in the range [0, 0.1]. We can observe that the cross polarized component is small, even at its maximum. Except for the existence of the cross polarized component, this situation is very similar to „external reflection" in the usual dielectric-dielectric case.

Case 2)

The constitutive parameters satisfy the relation

$$\frac{\sqrt{\mu\varepsilon}}{1+f} < \sqrt{\mu_0\varepsilon_0} < \frac{\sqrt{\mu\varepsilon}}{1-f} \tag{16}$$

which implies that for every angle of incidence there is always a real angle ϑ_1 of refraction corresponding to the left circularly polarized transmitted wave. But when the angle of incidence is greater than an angle ϑ_{0R}, given by

$$\sin\vartheta_{0R} = \left(\frac{\mu\varepsilon}{\mu_0\varepsilon_0}\right)^{1/2} \frac{1}{1+f} \tag{17}$$

the right circularly polarized transmitted wave becomes evanescent and propagates parallel to the interface.

Case 3)

In this case

$$\frac{\sqrt{\mu\varepsilon}}{1-f} < \sqrt{\mu_0\varepsilon_0} \tag{18}$$

and besides the angle ϑ_{0R}, there is also an angle of incidence ϑ_{0L}, satisfying

$$\sin\vartheta_{0L} = \left(\frac{\mu\varepsilon}{\mu_0\varepsilon_0}\right)^{1/2} \frac{1}{1-f} \tag{19}$$

such that the left circularly polarized transmitted wave becomes evanescent when $\vartheta_0 > \vartheta_{0L}$.

Cases 2 and 3 are the more interesting since novel features appear in the behavior of the complex reflection coefficients. The situation resembles the „internal reflection" case for achiral materials, where total reflection takes place when the incidence angle is greater than the critical angle of total reflection. This is shown in fig. 2 where we have plotted the total reflectivity of the surface $|A_r^s|^2 + |A_r^p|^2$ when the incident wave is either S ($A_i^s = 1$) or P ($A_i^p = 1$) polarized. The constitutive parameters are $\varepsilon_0 = 1.33\,\varepsilon$, $\mu = \mu_0$ and $f = 0.09$ (case 3), for which case $\vartheta_{0R} = 52.60°$ and $\vartheta_{0L} = 72.12°$. We can observe that for incidences less than ϑ_{0L} both circular polarizations components are transmitted and the total reflectivity is very small, with jumps at ϑ_{0R} and ϑ_{0L}. When $\vartheta_{0R} < \vartheta_0 < \vartheta_{0L}$ only the left circularly polarized component is transmitted into the chiral material and when $\vartheta_0 > \vartheta_{0L}$ all the incident energy is reflected back into the achiral medium.

Graphs of $|A_r^s|^2$, $|A_r^p|^2$ and $|A_r^s/A_r^p|$ have been given in [5]. Comparing with case 1 where the small cross polarized component was the most sensitive to changes in f, in these cases each polarization exhibits notable values of θ_0 which are highly dependent on the chirality (β) of the medium. Moreover, the energy reflected back into the dielectric can be an important fraction of incident light. These facts show that experimental configurations corresponding to cases 2 and 3 could be particularly adequate to characterize chiral media by means of reflection ellipsometry.

In some experimental situations the phase of the reflected firelds plays a more important role than the amplitude, as for example in interferometric applications or in the study of non specular phenomena using bounded beams [11]. The phase of each coefficient has been plotted

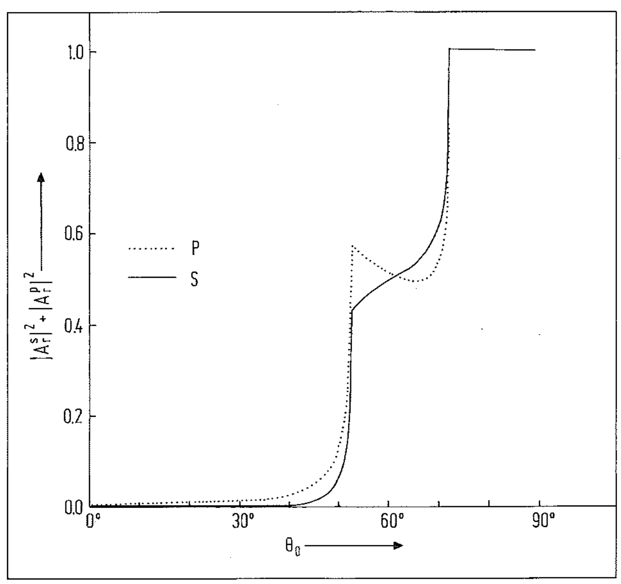

Fig. 2. Plot of $|A_r^s|^2 + |A_r^p|^2$ versus angle of incidence for $\mu = \mu_0$, $\varepsilon = 0.75\,\varepsilon_0$ and $f = 0.09$ for S and P polarized incidences.

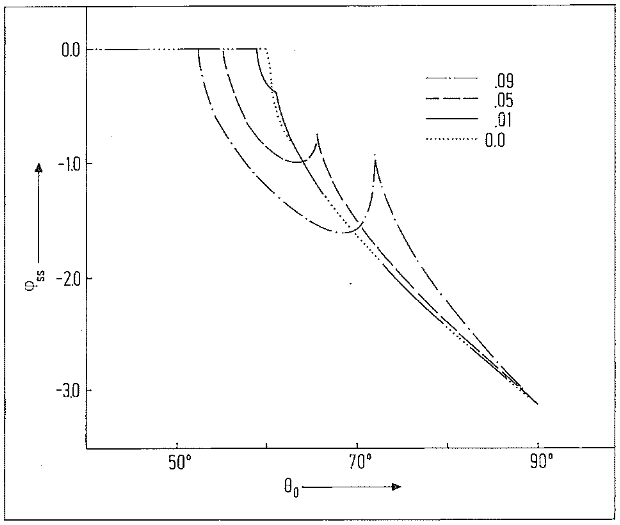

Fig. 3. **Plot of the phase of R_{ss} versus angle of incidence of $\mu = \mu_0$, $\varepsilon = 0.75\,\varepsilon_0$ and different values of f in the range [0, 0.1].**

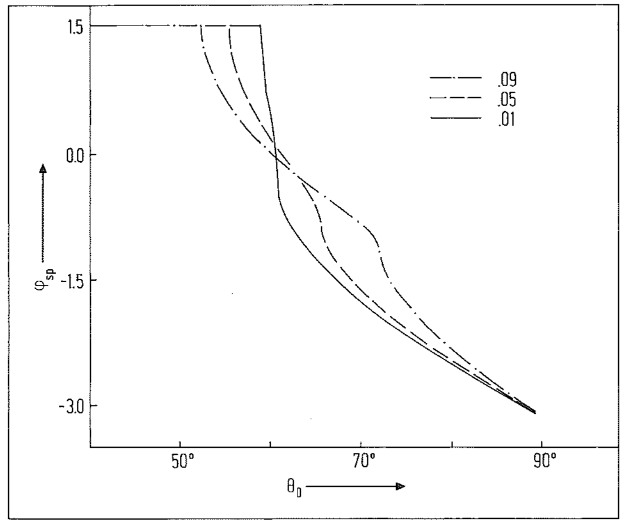

Fig. 5. **Plot of the phase of R_{sp} versus angle of incidence of $\mu = \mu_0$, $\varepsilon = 0.75\,\varepsilon_0$ and different values of f in the range [0, 0.1].**

in figs. 3–5 as functions of incidence for $\varepsilon_0 = 1.33\,\varepsilon$, $\mu = \mu_0$ with f as a parameter. The curves corresponding to an achiral interface ($f = 0$) are given as a reference. We found that the presence of chirality introduces new features in the phase curves. R_{ss} is in phase with the incident wave between normal incidence and ϑ_{0R} but from ϑ_{0R} up to grazing incidence the phase of R_{ss} (fig. 3) changes from zero to $-\pi$. The fact that either one or both circularly polarized transmitted components are evanescent, is responsible for different shapes of the curves in the regions $\vartheta_{0R} < \vartheta_0 < \vartheta_{0L}$ and $\vartheta_{0L} < \vartheta_0 < \pi/2$. The phase of R_{pp} (fig, 4) shows similar behavior except that it jumps from π to zero at $\vartheta_0 < \vartheta_{0R}$. The cross polarized reflected wave is always $\pi/2$ out of phase with respect to the incident wave whenever $\vartheta_0 < \vartheta_{0R}$ (see fig. 5). For incidences between ϑ_{0R} and $\pi/2$ the phase of R_{sp} changes continuously

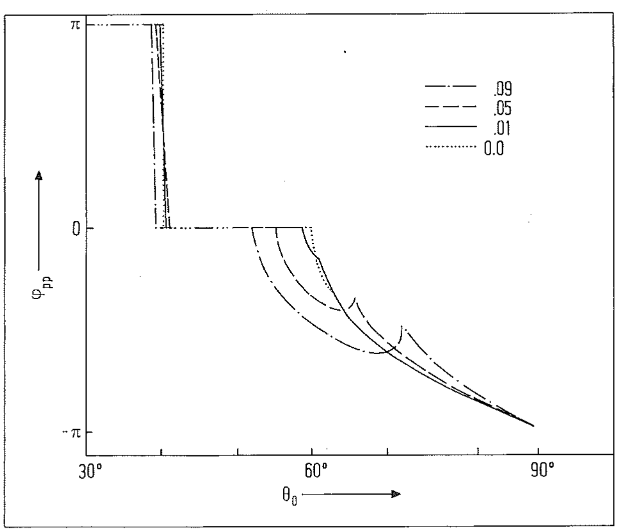

Fig. 4. **Plot of the phase of R_{pp} versus angle of incidence of $\mu = \mu_0$, $\varepsilon = 0.75\,\varepsilon_0$ and different values of f in the range [0, 0.1].**

from $\pi/2$ to $-\pi$ with an inflection point at ϑ_{0L}. These curves show that experimental configurations corresponding to cases 2 and 3 could be particularly significant to characterize chiral media by means of reflection ellipsometry.

4. Conclusion

In order to help to exploit all the possibilities of reflection ellipsometry as a tool to characterize chiral media, we have presented curves giving the complex reflection matrix elements as functions of angle of incidence and for different values of the constitutive parameters. Both magnitude and phase curves show noticeable features that could be exploited either to implement usual or novel ellipsometric techniques, or to predict the existence of novel manifestations of chiral asymmetries [6] which might deserve further attention. Of special interest in the study of the reflection of spatially limited beams are the regions where abrupt phase changes occur [11–12], since it is in these regions where non specular phenomena take place. For example, it is well known that bounded beams impinging at dielectric-dielectric interfaces at incidences near the critical angle of total reflection [13], suffer a lateral displacement intimately related with the value of the derivative of the phase with respect to angle of incidence. From the results presented here we can expect that bounded beams reflected at a chiral interface will exhibit not only a similar lateral shift at incidences near ϑ_{0R} but also an enhancement of this shift due to the sharp edge in the phase curves near ϑ_{0L}.

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