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Stationary property of normal-incidence reflection from isotropic surfaces

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The complex reflection coefficients for the parallel (p) and perpendicular (s) polarizations of light that are normally incident upon an isotropic surface are proved to be stationary with respect to small changes of the angle of incidence in the neighborhood of zero. This is true not only for a single interface between isotropic media but also for any one-dimensionally inhomogeneous or multilayer reflecting structure that is stratified in the direction of the surface normal. For incident light of certain intensity, phase, and polarization, the intensity, phase, and polarization of the reflected light all remain stationary with respect to small near-normal angle-of-incidence variations to first order. A second-order analysis is carried out to determine the parabolic (quadratic) variation of various reflection parameters of an interface with an angle near normal incidence.

1. INTRODUCTION

An established method for the determination of optical properties (complex refractive index or complex dielectric function) of materials (absorbing solids or liquids) is based on reflectance measured at normal incidence, usually as a function of incident photon energy or wavelength.¹ In carrying out such measurements, normal incidence is often loosely interpreted to mean incidence at angles within $\sim 15^\circ$ to the surface normal. The assumption is that Fresnel's reflection coefficients are stationary with respect to small angle-of-incidence variations near normal incidence. In this paper we prove that this stationary property indeed holds and that it is more general than is commonly thought. We find that it applies to (1) both the parallel (p) and perpendicular (s) polarizations (hence to all incident polarizations), (2) the magnitude and angle of the reflection coefficients (hence to the amplitude and phase of the reflected wave), and (3) a single interface as well as to multilayer or one-dimensional continuously inhomogeneous structures. Furthermore, the quadratic variations of reflection parameters of an interface with angle near normal incidence are determined.

2. REFLECTION BY A SINGLE INTERFACE

For a planar interface between two homogeneous and isotropic media of dielectric functions ϵ_0 and ϵ_1 , Fresnel's complex amplitude-reflection coefficients for the s and p polarizations at an angle of incidence ϕ (in medium 0) are given by²

$$r_{01s} = (S_0 - S_1)/(S_0 + S_1), \quad (1)$$

$$r_{01p} = (\epsilon_1 S_0 - \epsilon_0 S_1)/(\epsilon_1 S_0 + \epsilon_0 S_1), \quad (2)$$

where

$$S_i = (\epsilon_i - \epsilon_0 \sin^2 \phi)^{1/2}, \quad i = 0, 1. \quad (3)$$

If we take the first derivative with respect to ϕ (to be indicated throughout by a prime superscript) of Eqs. (1)–(3), we get

$$r_{01s}' = 2(S_0'S_1 - S_0S_1')/(S_0 + S_1)^2, \quad (4)$$

$$r_{01p}' = 2\epsilon_0\epsilon_1(S_0'S_1 - S_0S_1')/(\epsilon_1S_0 + \epsilon_0S_1)^2, \quad (5)$$

$$S_i' = a/S_i, \quad (6)$$

where

$$a = -\frac{1}{2}\epsilon_0 \sin 2\phi. \quad (7)$$

At normal incidence, $\phi = 0$, $\sin 2\phi = 0$, $a = 0$, and $S_i' = 0$. Consequently, from Eqs. (4) and (5), it follows also that

$$r_{01s}' = r_{01p}' = 0, \quad (8)$$

irrespective of ϵ_0 and ϵ_1 , i.e., for all interfaces. Equation (8) represents the stationary property of Fresnel's complex interface reflection coefficients at normal incidence.

3. REFLECTION BY A SINGLE-LAYER-COATED OR MULTILAYER-COATED SUBSTRATE

The complex amplitude-reflection coefficients of an all-isotropic single-layer-coated substrate are given by³

$$R_\nu = \frac{r_{01\nu} + r_{12\nu}X}{1 + r_{01\nu}r_{12\nu}X}, \quad \nu = p, s, \quad (9)$$

$$X = \exp[-j4\pi(d/\lambda)S_1], \quad (10)$$

where $r_{ij\nu}$ is Fresnel's reflection coefficient of the ij interface for the ν polarization; 0, 1, and 2 represent the medium of incidence (ambient), film, and substrate, respectively; d is the thickness of the homogeneous uniform layer, λ is the wavelength; and S_1 is as previously defined in Eqs. (3).

Differentiation of Eqs. (9) with respect to ϕ gives

$$\begin{aligned} \frac{R_\nu'}{R_\nu} = & \frac{(r_{01\nu}' + r_{12\nu}'X + r_{12\nu}X')}{(r_{01\nu} + r_{12\nu}X)} \\ & - \frac{(r_{01\nu}'r_{12\nu}X + r_{01\nu}r_{12\nu}'X + r_{01\nu}r_{12\nu}X')}{(1 + r_{01\nu}r_{12\nu}X)}, \end{aligned} \quad (11)$$

where, from Eq. (10),

$$X' = -j4\pi(d/\lambda)S_1'X. \quad (12)$$

At normal incidence, $r_{01\nu}' = r_{12\nu}' = 0$ for $\nu = p, s$, as was proved in Section 2. Also from Eqs. (6) and (7), $S_1' = 0$ at $\phi = 0$, and that makes $X' = 0$. It follows from Eq. (11) that

$$R_\nu' = 0, \quad \nu = p, s, \quad (13)$$

i.e., the overall (three-phase) reflection coefficients are stationary with ϕ at $\phi = 0$.

If a second layer is added, Eqs. (9) can be amended to apply in this case by substituting for $r_{12\nu}$ the reflection coefficient of the substructure that consists of the substrate (now medium 3) and the first film (medium 2). Because the latter reflection coefficient is stationary at $\phi = 0$, the overall reflection coefficient for the two-layer-substrate system will be stationary. By recurrence, this argument continues to hold for any number of isotropic layers, so that Eqs. (13) are satisfied by the corresponding overall reflection coefficients.

An inhomogeneous structure that is continuously graded (in the surface-normal direction) can be approximated to any desired degree of accuracy by a stack of layers of sufficient number. It follows that the complex coefficients of reflection R_ν from such a structure satisfy the stationary property $R_\nu' = 0$ at $\phi = 0$.

4. DERIVED STATIONARY PROPERTIES

As we have found, the complex reflection coefficients R_ν of an isotropic structure are stationary with respect to small variations of ϕ near $\phi = 0$. This means that not only the reflectance, $|R_\nu|$ or $|R_\nu|^2$, but also the reflection phase shifts, $\delta_\nu = \arg R_\nu$ (where $\nu = p$ or $\nu = s$), are stationary with respect to ϕ near $\phi = 0$. To see this, write any complex amplitude-reflection coefficient as

$$R_\nu = |R_\nu|e^{j\delta_\nu}. \quad (14)$$

Differentiation of Eq. (14) gives

$$\frac{R_\nu'}{R_\nu} = \frac{|R_\nu|'}{|R_\nu|} + j\delta_\nu'. \quad (15)$$

In Eq. (15), if we set $R_\nu' = 0$, we get

$$|R_\nu|' = 0, \quad (16)$$

$$\delta_\nu' = 0. \quad (17)$$

Equations (16) and (17) express the stationary behavior of the normal-incidence amplitude reflectance and phase shift, respectively, with respect to small ϕ changes. Of course, the intensity reflectance

$$\mathcal{R}_\nu = |R_\nu|^2 \quad (18)$$

is also stationary because

$$\mathcal{R}_\nu' = 2|R_\nu||R_\nu|' = 0 \quad (19)$$

when $|R_\nu|' = 0$.

Also, the (ellipsometric) ratio of p - and s -reflection coefficients (hence the state of polarization of the reflected light for a given incident polarization) is stationary with respect to small ϕ changes near $\phi = 0$. Thus if we write

$$\rho = R_p/R_s, \quad (20)$$

then

$$\rho' = (R_p'/R_s) - (R_p R_s'/R_s^2) \quad (21)$$

and

$$\rho' = 0 \quad (22)$$

when $R_p' = R_s' = 0$ at $\phi = 0$.

If an angle-of-incidence derivative ellipsometry and reflectometry experiment^{4,5} is carried out at normal incidence, all derivative signals will disappear to first order in the angle-of-incidence excursion $\Delta\phi$.

5. ANISOTROPIC SURFACES

Although details are not included here, it can be proved⁶ that the stationary property of normal-incidence reflection extends to uniaxial anisotropic stratified structures in which the optic axis coincides with the direction of stratification and is normal to the surface. We expect that such a property will also continue to hold for other symmetric anisotropies; however, further generalization cannot be made at this point.

6. SECOND DERIVATIVES FOR A SINGLE INTERFACE

Because the first derivatives with respect to ϕ of Fresnel's interface reflection coefficients are zero at $\phi = 0$, the change of these coefficients, as one goes off normal incidence by a small angle, is determined by the next nonvanishing higher-order derivative. Differentiation of Eqs. (4) and (5) gives the following second derivatives that are valid at any ϕ :

$$r_{01s}'' = [2(S_0 + S_1)(S_0''S_1 - S_0S_1'') - 4(S_0' + S_1')(S_0'S_1 - S_0S_1')]/(S_0 + S_1)^3, \quad (23)$$

$$r_{01p}'' = 2\epsilon_0\epsilon_1[(\epsilon_1S_0 + \epsilon_0S_1)(S_0''S_1 - S_0S_1'') - 2(\epsilon_1S_0' + \epsilon_0S_1')(S_0'S_1 - S_0S_1')]/(\epsilon_1S_0 + \epsilon_0S_1)^3, \quad (24)$$

where

$$S_i'' = (a'/S_i) - (a^2/S_i^3), \quad a' = -\epsilon_0 \cos 2\phi. \quad (25)$$

Evaluation of Eqs. (23) and (24) at $\phi = 0$ leads to the simple result that

$$r_{01s}''(0) = 2N_{01}r_{01s}(0), \quad (26)$$

$$r_{01p}''(0) = -2N_{01}r_{01p}(0), \quad (27)$$

where

$$N_{01} = (\epsilon_0/\epsilon_1)^{1/2} \quad (28)$$

is the relative refractive index of the interface and the argument 0 indicates that the functions are evaluated at $\phi = 0$. The difference in sign between Eqs. (26) and (27) is a consequence of our adoption of the Nebraska (Muller) conventions.⁷

It is now possible to write down the first three terms (the second term being zero) of a Maclaurin-series expansion,

$$f(x) = f(0) + xf'(0) + \frac{x^2}{2!}f''(0) + \dots,$$

of Fresnel's interface reflection coefficients near normal incidence:

$$r_{01s}''(\phi) = r_{01s}(0)(1 + N_{01}\phi^2 + \dots), \quad (29)$$

$$r_{01p}''(\phi) = r_{01p}(0)(1 - N_{01}\phi^2 + \dots). \quad (30)$$

From Eqs. (29) and (30) the ratio of reflection coefficients $\rho = r_{01p}/r_{01s}$ is

$$\rho(\phi) = \rho(0)(1 - 2N_{01}\phi^2 + \dots), \quad (31)$$

where $\rho(0) = -1$ in the Nebraska (Muller) conventions.⁷

It is also convenient to work with the fractional changes of Fresnel's coefficients and their ratio that result from a change of angle of incidence from zero (normal incidence) to a small angle ϕ :

$$\Delta r_{01\nu}/r_{01\nu} = \pm N_{01}\phi^2, \quad (32)$$

$$\Delta\rho/\rho = -2N_{01}\phi^2. \quad (33)$$

In Eq. (32), the plus and the minus correspond to $\nu = s$ and $\nu = p$, respectively. Writing $r = |r|e^{j\delta}$, $\rho = |\rho|e^{j\Delta}$, and taking the real and imaginary parts of Eqs. (32) and (33), we obtain

$$\Delta|r_{01\nu}|/|r_{01\nu}| = \pm n_{01}\phi^2, \quad (34)$$

$$\Delta\delta_{01\nu} = \pm k_{01}\phi^2, \quad (35)$$

$$\Delta|\rho|/|\rho| = -2n_{01}\phi^2, \quad (36)$$

$$\Delta\Delta = -2k_{01}\phi^2. \quad (37)$$

It also follows from Eqs. (18) and (34) that the fractional change of intensity reflectance is given by

$$\Delta\mathcal{R}_{01\nu}/\mathcal{R}_{01\nu} = \pm 2n_{01}\phi^2. \quad (38)$$

In Eqs. (34), (35), and (38), the upper and lower signs correspond to $\nu = s$ and $\nu = p$, respectively:

$$N_{01} = n_{01} + jk_{01} \quad (39)$$

and

$$N_{01} = 1/N_{10} = 1/(n_{10} - jk_{10}) = (n_{10} + jk_{10})/(n_{10}^2 + k_{10}^2). \quad (40)$$

When the medium of incidence is vacuum or air ($N_0 = 1$), n_{10} and k_{10} represent the refractive index and extinction coefficient of the medium of refraction, respectively. Equations (34)–(38) determine the ranges of angle of incidence over which the different reflection parameters change by specified amounts from their normal-incidence values. For instance, Eq. (38) can be solved for ϕ :

$$\begin{aligned} \phi &= |\Delta\mathcal{R}/\mathcal{R}|^{1/2}/(2n_{01})^{1/2} \\ &= |\Delta\mathcal{R}/\mathcal{R}|^{1/2}(n_{10}^2 + k_{10}^2)^{1/2}/(2n_{10})^{1/2}. \end{aligned} \quad (41)$$

For an air–glass interface, $n_{10} = 1.5$, $k_{10} = 0$, and we get $\phi = 5^\circ$, 10° for 1 and 4% fractional changes in reflectance, respectively. For an air–gold interface at $\lambda = 0.55 \mu\text{m}$, $n_{10} = 0.33$, $k_{10} = 2.32$ (Ref. 8), and the ranges of ϕ , calculated from Eq. (41), for $\Delta\mathcal{R}/\mathcal{R} = 1, 4\%$ are 16.5° and 33° , respectively. For transparent interfaces, $k_{10} = k_{01} = 0$, Eqs. (35) and (37) indicate that all phase changes are zero to second order in ϕ . This is in agreement with the fact that all reflection phase shifts stay exactly constant up to the Brewster angle, with δ_p and Δ experiencing discontinuous jumps of π at that angle. The phase changes at the air–gold interface in going from $\phi = 0$ to $\phi = 15^\circ$ are $\Delta\delta_s = -\Delta\delta_p = 1.66^\circ$ and $\Delta\Delta = -3.32^\circ$, as calculated from Eqs. (35) and (37), respectively.

It is possible to extend the second-order analysis to an arbitrary multilayer structure. A specific example of such an analysis, for high-reflectance multilayer dielectric mirrors, has been published previously.⁹

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