Reflection at oblique incidence and the existence of a Brewster angle

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We show that when the ratio r_p/r_s of the reflection amplitudes for the electromagnetic p and s waves is taken to be 1 at normal incidence, it will have the value -1 at grazing incidence. This result is valid for sharp or diffuse interfacial profiles, for internal as well as external reflections, and in the presence of absorption and anisotropy within the reflecting layer or its substrate. (The anisotropy of the dielectric function is limited to a difference in the response of the system to electric fields perpendicular or parallel to the interface, characterized by ϵ_{\perp} and ϵ_{\parallel} .) Under these conditions, there will always be at least one angle of incidence at which the real part of r_p/r_s is zero. Under the same conditions, the reflected s and p electric fields at grazing incidence are out of phase with the incident electric fields, thus producing destructive interference at the mirror's edge in Lloyd's mirror experiment.

1. INTRODUCTION

The existence and location of a Brewster angle are of importance in ellipsometry and in particular in the application of the polarization-modulation ellipsometric technique of Jasperson and Schnatterly¹ to the study of liquid surfaces.² There the quantity most easily measured is $\overline{\rho}$, the value of the imaginary part of the ratio of the p and s reflection amplitudes at the angle where the real part is zero. The angle at which $\operatorname{Re}(r_p/r_s) = 0$ is one of several possible operational definitions of the Brewster angle.³ In this paper we show that, under rather general conditions, at least one angle of incidence will exist at which $\operatorname{Re}(r_p/r_s) = 0$. We first establish that, provided that the response of the planar system is independent of the azimuthal angle, if $r_p/r_s = 1$ at normal incidence, then r_p/r_s = -1 at grazing incidence. The existence of an angle at which $\operatorname{Re}(r_p/r_s) = 0$ then follows from continuity. An interesting result obtained en route is that r_p and r_s take the values +1 and -1 at grazing incidence, exactly and without ambiguity of phase.

2. THE s-WAVE REFLECTION AMPLITUDE

We consider plane electromagnetic waves incident upon an interface lying in the xy plane. When the propagation is in the zx plane, $\mathbf{E} = (0, E_y, 0)$ for the s wave and E_y satisfies⁴

$$\nabla^2 E_y + \epsilon \frac{\omega^2}{c^2} E_y = 0, \qquad (1)$$

where c is the speed of light and ω is the angular frequency of the (monochromatic) wave. When ϵ , the dielectric function, is assumed to be a function of z only, $E_y = \exp(iKx)E(z)$, where E(z) satisfies

$$\frac{\mathrm{d}^2 E}{\mathrm{d}z^2} + \left(\epsilon \frac{\omega^2}{c^2} - K^2\right) E = 0. \tag{2}$$

K is the x component of the wave vector in either medium, so

if θ_1 and θ_2 are the angles of incidence and refraction, $K = (\epsilon_1)^{1/2} (\omega/c) \sin \theta_1 = (\epsilon_2)^{1/2} (\omega/c) \sin \theta_2$, where ϵ_1 and ϵ_2 are the limiting values of $\epsilon(z)$ at $-\infty$ and $+\infty$. The quantity

$$q^{2}(z) = \epsilon(z) \frac{\omega^{2}}{c^{2}} - K^{2}$$
(3)

is the square of the wave-number component perpendicular to the interface; q(z) takes the limiting values $q_1 = \sqrt{\epsilon_1(\omega/c)\cos\theta_1}$ and $q_2 = \sqrt{\epsilon_2(\omega/c)\cos\theta_2}$. E(z) has the asymptotic forms

$$\exp(iq_1z) + r_s \exp(-iq_1z) \leftarrow E(z) \rightarrow t_s \exp(iq_2z).$$
(4)

This equation defines the reflection and transmission amplitudes r_s and t_s .

We now consider interfaces for which $\epsilon = \epsilon_1$ for $z < z_1$ and $\epsilon = \epsilon_2$ for $z > z_2$; the thickness $z_2 - z_1$ of the nonuniform region can be large. The prescription includes, by a limiting process, the dielectric functions used in diffuse fluid-fluid interfaces, such as

$$\epsilon(z) = \frac{1}{2}(\epsilon_1 + \epsilon_2) - \frac{1}{2}(\epsilon_1 - \epsilon_2) \tanh[(z - z_0)/2a].$$
(5)

In the example given, one could take $z_1 - z_0 = -\Delta z/2$, $z_2 - z_0 = \Delta z/2$, and by making $\Delta z/a$ large enough, any desired accuracy can be achieved. Now Eq. (2) is a second-order linear differential equation and thus has two linearly independent solutions [for an arbitrary form of $\epsilon(z)$]. We call these A(z) and B(z) in the region $z_1 \leq z \leq z_2$. Then

$$E(z) = \begin{cases} \exp(iq_1 z) + r_s \exp(-iq_1 z), & z < z_1 \\ \alpha A(z) + \beta B(z), & z_1 \le z \le z_2 \\ t_s \exp(iq_2 z), & z > z_2 \end{cases}$$
(6)

The continuity of E and dE/dz at z_1 and z_2 gives us four equations in the four unknown coefficients r_s , t_s , α , and β . Solving for r_s , we find⁵ (writing A_1 for $A(z_1)$, A_1' for dA/dz at z_1 , etc.) that John Lekner

$$r_{s} = \exp(2iq_{1}z_{1})\frac{q_{1}q_{2}(A_{1}B_{2} - B_{1}A_{2}) + iq_{1}(A_{1}B_{2}' - B_{1}A_{2}') + iq_{2}(A_{1}'B_{2} - B_{1}'A_{2}) - (A_{1}'B_{2}' - B_{1}'A_{2}')}{q_{1}q_{2}(A_{1}B_{2} - B_{1}A_{2}) + iq_{1}(A_{1}B_{2}' - B_{1}A_{2}') - iq_{2}(A_{1}'B_{2} - B_{1}'A_{2}) + (A_{1}'B_{2}' - B_{1}'A_{2}')}$$
(7)

The result that $r_s \rightarrow -1$ for grazing incidence follows immediately on letting $q_1 = \sqrt{\epsilon_1}(\omega/c)\cos\theta_1 \rightarrow 0$. It also follows easily that the reflection amplitude for an arbitrary nonsingular profile shape of extent Δz approaches the step or sharp interface value as Δz tends to zero⁵:

$$r_s \to \exp(2iq_1z_1) \frac{q_1 - q_2}{q_1 + q_2}$$
 as $\Delta z \to 0.$ (8)

Note that even in the limit of a sharp transition from ϵ_1 to ϵ_2 , there is an arbitrariness in the phase of the reflection amplitude (associated with the arbitrariness of the location of the step relative to the origin). However, at grazing incidence, when $q_1 \rightarrow 0$, this arbitrariness disappears, and the reflection amplitude is known in magnitude and in phase. The incident and reflected waves are then moving parallel to the interface, and there is no motion perpendicular to the interface to give rise to a phase shift associated with the path difference $2z_1$ between the incident and reflected waves.

We note also that the electric field is reversed on reflection at grazing incidence under *all* conditions (including total interal reflection). This is a general property of waves satisfying equations of the form $d^2\psi/dz^2 + q^2\psi = 0$. For example, nonrelativistic quantum particles of mass *m* and energy *E*, moving in a potential V(z), satisfy a Schrödinger equation in which the *z* variation has this form, with $q^2(z) = (2m/\hbar^2)[E$ $- V(z)] - K^2$. Thus we have proved that, at grazing incidence, the reflected probability amplitude for electrons, neutrons, etc., will be equal in magnitude to, and out of phase with, the incident probability amplitude.

3. THE *p*-WAVE REFLECTION AMPLITUDE

We again take the incident and reflected waves propagating in the zx plane and the interface lying in the xy plane. For the p wave, $\mathbf{B} = (0, B_y, 0), B_y = \exp(iKx)B(z)$ (when ϵ is a function of z only), and B(z) satisfies⁴

$$-\frac{\sin\theta_1}{\sqrt{\epsilon_1}}\exp(iKx)[\exp(iq_1z) - r_p\,\exp(-iq_1z)]$$

$$\leftarrow E_z \rightarrow \frac{-\sin\theta_2}{\sqrt{\epsilon_1}}t_p\,\exp(iKx + iq_2z). \quad (12)$$

The reflection amplitude is defined as the ratio of the coefficient of $\exp(-iq_1z)$ to the coefficient of $\exp(iq_1z)$. We see that the reflection amplitudes for E_x and E_z (the electric-field components parallel and perpendicular to the interface) have opposite sign. At normal incidence, there is no physical difference between the s and p waves. E_z is then zero, and (for our definition of r_p and t_p) $r_p = r_s$, $t_p = t_s$. The opposite convention (with $r_p = -r_s$ at normal incidence) is also in use.^{6,7}

At normal incidence (K = 0), the Maxwell equation $\nabla \times \mathbf{E} = -(1/c)\partial \mathbf{B}/\partial t$ gives $\partial E_x/\partial z = i(\omega/c)B_y$; thus *B*, the solution of Eqs. (9) and (10), must be proportional to dE/dz, where *E* is the solution of Eqs. (2) and (4). This is indeed the case, as may be verified by substituting dE/dz for *B* in Eq. (9) and using Eq. (2).

We will now derive a general expression for r_p , analogous to the result of Eq. (7) for r_s . Let C(z) and D(z) be two linearly independent solutions of Eq. (9) within the interval (z_1, z_2) . Then

$$B(z) = \begin{cases} \exp(iq_{1}z) - r_{p} \exp(-iq_{1}z), & z < z_{1} \\ \gamma C(z) + \delta D(z), & z_{1} \le z \le z_{2} \\ \left(\frac{\epsilon_{2}}{\epsilon_{1}}\right)^{1/2} t_{p} \exp(iq_{2}z), & z > z_{2} \end{cases}$$
(13)

The form of Eq. (9) shows that $dB/\epsilon dz$ must be continuous (discontinuity in $dB/\epsilon dz$ would give rise to a delta-function term). On using the continuity of B and $dB/\epsilon dz$ at z_1 and z_2 , we obtain four equations in the four unknowns r_p , t_p , γ , δ . Solving for r_p , we find that

$$r_{p} = -\exp(2iq_{1}z_{1})\frac{Q_{1}Q_{2}(C_{1}D_{2} - D_{1}C_{2}) + iQ_{1}(C_{1}D_{2}' - D_{1}C_{2}') + iQ_{2}(C_{1}'D_{2} - D_{1}'C_{2}) - (C_{1}'D_{2}' - D_{1}'C_{2}')}{Q_{1}Q_{2}(C_{1}D_{2} - D_{1}C_{2}) + iQ_{1}(C_{1}D_{2}' - D_{1}C_{2}') - iQ_{2}(C_{1}'D_{2} - D_{1}'C_{2}) + (C_{1}'D_{2}' - D_{1}'C_{2}')},$$
(14)

$$\frac{\mathrm{d}}{\mathrm{d}z}\left(\frac{1}{\epsilon}\frac{\mathrm{d}B}{\mathrm{d}z}\right) + \left(\frac{\omega^2}{c^2} - \frac{K^2}{\epsilon}\right)B = 0. \tag{9}$$

We take the asymptotic forms of B(z) to be³

$$\exp(iq_1z) - r_p \exp(-iq_1z) \leftarrow B(z) \rightarrow \left(\frac{\epsilon_2}{\epsilon_1}\right)^{1/2} t_p \exp(iq_2z).$$
(10)

The reason for the factors -1 and $(\epsilon_1/\epsilon_1)^{1/2}$ multiplying r_p and t_p is that we wish r_s and r_p , and t_s and t_p , to refer to the same quantity (here chosen to be the electric field) and to be equal to normal incidence. The electric-field components of the p wave are found from $\mathbf{E} = (ic/\epsilon\omega)\nabla \times \mathbf{B}$, the time-harmonic consequence of $\nabla \times \mathbf{B} = (\epsilon/c)\partial \mathbf{E}/\partial t$. From Eq. (10) we find that

$$\frac{\cos\theta_1}{\sqrt{\epsilon_1}} \exp(iKx) [\exp(iq_1 z) + r_p \exp(-iq_1 z)]$$

$$\leftarrow E_x \rightarrow \frac{\cos\theta_2}{\sqrt{\epsilon_1}} t_p \exp(iKx + iq_2 z), \quad (11)$$

where C_1 denotes $C(z_1)$, C_1' denotes dC/dz at z_1 , etc., and $Q_1 = q_1/\epsilon_1$, $Q_2 = q_2/\epsilon_2$. On setting $Q_1 = 0$, we find that $r_p \rightarrow 1$ at grazing incidence; the method used in Ref. 5 to prove Eq. (8) gives

$$r_p \rightarrow -\exp(2iq_1z_1) \frac{Q_1 - Q_2}{Q_1 + Q_2}$$
 as $\Delta z \rightarrow 0.$ (15)

The fact that $r_p \rightarrow 1$ at grazing incidence shows, together with Eq. (12), that the electric field of the *p* wave is reversed by reflection. That the electric field of the *s* wave is reversed at grazing incidence was shown in Section 2. These results hold whether the reflecting surface is metallic or dielectric, sharp or diffuse, for internal as well as external reflection, and (as we show in Section 4) in the presence of anisotropy. It follows that the Lloyd mirror experiment should produce diffraction fringes, with destructive interference at the mirror's edge, under these general conditions. This is in accord with experiment.⁸

THE EFFECT OF ANISOTROPY WITHIN 4. THE INTERFACE AND THE SUBSTRATE

In all real interfaces, even single-component monatomic crystal-gas and liquid-gas systems,⁹ the dielectric response of the system is in principle different for the electric-field vector perpendicular and parallel to the interface. Two dielectric functions, $\epsilon_{\perp}(z)$ and $\epsilon_{\parallel}(z)$, thus enter Maxwell's equations; the s- and p-wave equations now become (Ref. 10, App. A)

 $\frac{\mathrm{d}^2 E}{\mathrm{d}z^2} + \left(\epsilon_{\parallel}\frac{\omega^2}{c^2} - K^2\right)E = 0$

and

$$\frac{\mathrm{d}}{\mathrm{d}z} \left(\frac{1}{\epsilon_{\parallel}} \frac{\mathrm{d}B}{\mathrm{d}z} \right) + \left(\frac{\omega^2}{c^2} - \frac{K^2}{\epsilon_{\perp}} \right) B = 0.$$
(17)

We again have the result that, at normal incidence, B =(constant) dE/dz. The s-wave equation has the same form as before, with ϵ_{\parallel} replacing ϵ and $q_E^2 = \epsilon_{\parallel}(\omega^2/c^2) - K^2$; the previous results thus follow. The p-wave equation now contains both ϵ_{\parallel} and ϵ_{\perp} ; outside the interfacial region the effective q_B^2 is $\epsilon_{\parallel}(\omega^2/c^2) - (\epsilon_{\parallel}/\epsilon_{\perp})K^2$. Provided that q_B and q_E are equal inside medium 1 (which must therefore be isotropic). the meaning of q_1 is the same for the s and p waves. The derivation of Eq. (14) proceeds as before [with C and D now the solutions of Eq. (17)]. Thus the results that $r_p \rightarrow 1$ and $r_s \rightarrow -1$ at grazing incidence remain valid in the presence of anisotropy.

The average electrodynamic properties of many systems are fully characterized by ϵ_{\parallel} and ϵ_{\perp} , even when there is molecular orientation at the interface, provided that the orientation is relative to the normal to the interface. When, however, there is alignment along a direction parallel to the interface, as can be the case in some liquid crystals, the system has lost azimuthal symmetry, and the description of reflection in terms of s and p waves is no longer adequate.

5. EXISTENCE OF A BREWSTER ANGLE

We have shown that when $r_p/r_s = 1$ at normal incidence, r_p/r_s \rightarrow -1 at grazing incidence. In the polarization-modulation ellipsometry technique, the angle of incidence for which $\operatorname{Re}(r_p/r_s) = 0$ is the operational definition of the Brewster

angle. Since r_p/r_s moves in the complex plane from the point +1 at normal incidence to -1 at grazing incidence, it follows that it must cross the line $\operatorname{Re}(r_p/r_s) = 0$ at least once (and in general an odd number of times). This is a consequence of the continuity of solutions of linear differential equations as a function of the parameters of the equations (see, for example, Ref. 11, in particular, Secs. 4 and 10 of Chap. 6).

The existence of at least one Brewster angle as defined above is thus established for all planar reflecting systems for which the s- and p-wave characterization is adequate. The presence of absorption is implicitly accounted for: We have not made the assumption that the dielectric functions of the interface or substrate are real. Anisotropy has been shown not to affect the main results, provided that the azimuthal symmetry remains unbroken.

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