Reflection of light by a nonuniform film between like media

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We derive variational expressions for the s- and p-wave reflection amplitudes at a nonuniform (and possibly absorbing) planar layer between like media, for example, a soap film in air. These variational expressions are correct at grazing incidence, in contrast to first-order perturbation-theory reflection amplitudes, which diverge there. The variational reflection amplitudes are also correct to second order in the ratio of the film thickness to the wavelength of the incident wave. The results for an anisotropic film are also given.

1. INTRODUCTION

Charmet and de Gennes[1] have recently derived ellipsometric formulas for reflection by an inhomogeneous layer bounded by a uniform dielectric (liquids or liquid mixtures bounded by glass are examples of practical interest[1,2]). Their method was perturbation theory, analogous to the Born approximation in scattering theory.[3] As we shall see below, the corresponding perturbation-theory results for reflection by an inhomogeneous layer between like media fail at grazing incidence: the first-order perturbation reflection amplitudes diverge there. (For passive media, the reflection amplitudes must not go outside the unit circle.) To deal with this problem, we have adapted Schwinger's variational method of scattering theory[4,5] to the reflection problem. With the same input as first-order perturbation theory (the plane wave of the first-order Born approximation), the variational reflection amplitudes are correct at grazing incidence and are further exact to second order in the ratio of film thickness to wavelength. These results are derived in the next three sections; comparison of $|r_s|^2$, $|r_p|^2$, and r_p/r_s with the exact results for a simple model is made in Section 5. The effect of anisotropy is considered in Section 6. To aid in the derivation of the formulas in the anisotropic case, a first-principles derivation of the equations satisfied by the s and p waves is given in Sections 2 and 3.

2. s-WAVE REFLECTION AMPLITUDE

We consider plane electromagnetic waves incident upon a film lying in the xy plane and characterized by a dielectric-function profile, $\epsilon(z)$. The media on either side of the film have $\epsilon = \epsilon_0$, a constant. When the propagation is in the zx plane, $\mathbf{E} = (0, E_y, 0)$, and for monochromatic waves of angular frequency ω [with time dependence $\exp(-i\omega t)$], the Maxwell equation $\nabla \times \mathbf{E} = -(1/c)\partial\mathbf{B}/\partial t$ gives

$$-\frac{\partial E_y}{\partial z} = i\frac{\omega}{c}B_x, \qquad \frac{\partial E_y}{\partial x} = i\frac{\omega}{c}B_z \tag{1}$$

and $B_y = 0$. (c is the speed of light.) The complementary equation $\nabla \times \mathbf{B} = (\epsilon/c)\partial\mathbf{E}/\partial t$ gives

$$\frac{\partial B_x}{\partial z} - \frac{\partial B_z}{\partial x} = -i\epsilon\frac{\omega}{c}E_y. \tag{2}$$

On eliminating B_x and B_z from Eqs. (1) and (2) we obtain a second-order partial differential equation for E_y,

$$\frac{\partial^2 E_y}{\partial z^2} + \frac{\partial^2 E_y}{\partial x^2} + \epsilon\frac{\omega^2}{c^2}E_y = 0. \tag{3}$$

Because $\epsilon = \epsilon(z)$, we may write

$$E_y(z, x) = \exp(iKx)E(z), \tag{4}$$

where $E(z)$ satisfies

$$\frac{\mathrm{d}^2E}{\mathrm{d}z^2} + q^2E = 0, \qquad q^2(z) = \epsilon(z)\frac{\omega^2}{c^2} - K^2. \tag{5}$$

The separation of variables constant K is the component of the wave vector along the interface. Thus

$$K = \sqrt{\epsilon_0}\frac{\omega}{c}\sin\theta, \tag{6}$$

where θ is the angle of incidence. The component perpendicular to the interface is $q(z)$ and takes the limiting value

$$q_0 = \sqrt{\epsilon_0}\frac{\omega}{c}\cos\theta, \tag{7}$$

within the uniform medium on either side of the film.

The reflection amplitude r_s and transmission amplitude t_s are defined in terms of the asymptotic forms of the solution of Eqs. (5):

$$\exp(iq_0z) + r_s\exp(-iq_0z) \leftarrow E \rightarrow t_s\exp(iq_0z). \tag{8}$$

One constructs a perturbation theory for r_s in terms of the solution $E_0(z) = \exp(iq_0z)$ for the case where $\epsilon = \epsilon_0$ everywhere. This is done by means of a Green function $G(z, \zeta)$ satisfying

$$\frac{\partial^2 G}{\partial z^2} + q_0^2G = \delta(z - \zeta). \tag{9}$$

We can then write an integral form of Eqs. (5) in terms of E_0 and G as

$$E(z) = E_0(z) - \int_{-\infty}^{\infty}\mathrm{d}\zeta\Delta q^2(\zeta)G(z, \zeta)E(\zeta). \tag{10}$$

Iteration of Eq. (10) gives successive orders [in $\Delta q^2 = q^2 - q_0^2 = (\epsilon - \epsilon_0)\omega^2/c^2$] in the expansion $E = E_0 + E_1 + \ldots$. To first order in Δq^2,

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$$E_1(z) = -\int_{-\infty}^{\infty} d\zeta \Delta q^2(\zeta) G(z, \zeta) E_0(\zeta). \quad (11)$$

The Green function appropriate to our problem is[6]

$$G_s(z, \zeta) = \frac{\exp(iq_0|z - \zeta|)}{2iq_0}. \quad (12)$$

The first-order perturbation value for the reflection amplitude is obtained from Eq. (11) by taking the limit $z \to -\infty$ and extracting the coefficient of $\exp(iq_0 z)$:

$$r_s^{\text{pert}} = -\frac{1}{2iq_0}\int_{-\infty}^{\infty} d\zeta \Delta q^2(\zeta)\exp(2iq_0\zeta). \quad (13)$$

Note that it diverges at grazing incidence (as $q_0 \to 0$).

We now adapt Schwinger's variational method for the scattering problem[4,5] to the reflection problem. We rewrite Eq. (10) as

$$E(z) + \int_{-\infty}^{\infty} d\zeta \Delta q^2(\zeta) E(\zeta) G(z, \zeta) = E_0(z), \quad (14)$$

multiply through by $\Delta q^2(z)E(z)$, and integrate over all z:

$$\int_{-\infty}^{\infty} dz \Delta q^2(z) E^2(z) + \int_{-\infty}^{\infty} dz \Delta q^2(z) E(z) \int_{-\infty}^{\infty} d\zeta \Delta q^2(\zeta) E(\zeta) G(z, \zeta) = \int_{-\infty}^{\infty} dz \Delta q^2(z) E(z) E_0(z). \quad (15)$$

We write this as $S = F$, where S (the left-hand side) is of second degree in E and F (the right-hand side) is of first degree in E. F is proportional to the reflection amplitude, as we see by extracting the asymptotic form of $E(z)$ from Eq. (10) as $z \to -\infty$; this is

$$\exp(iq_0 z) - \exp(-iq_0 z)\frac{1}{2iq_0}\int_{-\infty}^{\infty} d\zeta \Delta q^2(\zeta) E(\zeta) E_0(\zeta). \quad (16)$$

Comparison of expressions (8) and (16) shows that the exact reflection amplitude is

$$r_s = -F/2iq_0. \quad (17)$$

The variational principle for r_s is obtained by considering the shifts δS and δF as $E(z)$ is shifted by $\delta E(z)$: These are

$$\delta F = \int_{-\infty}^{\infty} dz \delta E(z) \Delta q^2(z) E_0(z) \quad (18)$$

and

$$\delta S = 2\int_{-\infty}^{\infty} dz \Delta q^2(z) \delta E(z)\left\{E(z) + \int_{-\infty}^{\infty} d\zeta \Delta q^2(\zeta) E(\zeta) G(z, \zeta)\right\}. \quad (19)$$

The expression inside the braces is $E_0(z)$, by Eq. (14), so $\delta S = 2\delta F$. But $S = F$, so $\delta S/S = 2\delta F/F$, or

$$\delta(F^2/S) = 0. \quad (20)$$

This is the variational principle: the correct E will extremize F^2/S. Using Eq. (17) we thus have a variational expression for r_s:

$$r_s^{\text{var}} = \frac{-F^2/S}{2iq_0}. \quad (21)$$

The simplest variational trial function for $E(z)$ is $E_0(z) = \exp(iq_0 z)$. This gives the values F_0 and S_0 for F and S, where

$$F_0 = \int_{-\infty}^{\infty} dz \Delta q^2(z) \exp(2iq_0 z) = -2iq_0 r_s^{\text{pert}} \quad (22)$$

[from Eq. (13)] and

$$S_0 = F_0 + \frac{1}{2iq_0}\int_{-\infty}^{\infty} dz \Delta q^2(z)\exp(iq_0 z)\int_{-\infty}^{\infty} d\zeta \Delta q^2(\zeta) \times \exp(iq_0\zeta)\exp(iq_0|z - \zeta|). \quad (23)$$

The corresponding variational estimate for r_s is

$$r_s^{\text{var}} = \frac{r_s^{\text{pert}}}{1 + (4q_0^2 r_s^{\text{pert}})^{-1}\int_{-\infty}^{\infty} dz \Delta q^2(z)\left\{\exp(2iq_0 z)\int_{-\infty}^{z} d\zeta \Delta q^2(\zeta) + \int_{z}^{\infty} d\zeta \Delta q^2(\zeta)\exp(2iq_0\zeta)\right\}}. \quad (24)$$

At grazing incidence ($q_0 \to 0$) this variational expression tends to -1, as is correct for any dielectric-function profile.[7] Further, Eq. (24) is correct to second order in the film thickness, as can be seen by comparing it with the expansion [Ref. 8, Eqs. (40) and (42)]

$$r_s = \frac{i}{2q_0}\int_{-\infty}^{\infty} dz \Delta q^2(z) - \int_{-\infty}^{\infty} dz \Delta q^2(z) z + \left\{\frac{i}{2q_0}\int_{-\infty}^{\infty} dz \Delta q^2(z)\right\}^2 + \ldots. \quad (25)$$

3. p-WAVE REFLECTION AMPLITUDE

We again take the incident and reflected waves propagating in the zx plane and the film lying in the xy plane. For the p wave, $\mathbf{B} = (0, B_y, 0)$; the Maxwell equation $\nabla \times \mathbf{B} = (\epsilon/c)\partial\mathbf{E}/\partial t$ gives $E_y = 0$ and

$$\frac{\partial B_y}{\partial z} = i\epsilon\frac{\omega}{c}E_x, \qquad \frac{\partial B_y}{\partial x} = -i\epsilon\frac{\omega}{c}E_z. \quad (26)$$

The complementary equation $\nabla \times \mathbf{E} = -(1/c)\partial\mathbf{B}/\partial t$ gives

$$\frac{\partial E_x}{\partial z} - \frac{\partial E_z}{\partial x} = i\frac{\omega}{c}B_y. \quad (27)$$

Elimination of E_x and E_z gives

$$\frac{\partial}{\partial z}\left(\frac{1}{\epsilon}\frac{\partial B_y}{\partial z}\right) + \frac{\partial}{\partial x}\left(\frac{1}{\epsilon}\frac{\partial B_y}{\partial x}\right) + \frac{\omega^2}{c^2}B_y = 0. \quad (28)$$

Since ϵ is a function of z only, we may write

$$B_y(z, x) = \exp(iKx)B(z), \quad (29)$$

where K has the same meaning as for the s wave and B satisfies

$$\frac{d}{dz}\left(v\frac{dB}{dz}\right) + \left(\frac{\omega^2}{c^2} - vK^2\right)B = 0, \qquad v = 1/\epsilon \quad (30)$$

and has the asymptotic forms

$$\exp(iq_0 z) - r_p\exp(-iq_0 z) \leftarrow B \rightarrow t_p\exp(iq_0 z). \quad (31)$$

(The reason for the minus in front of r_p is that we wish r_s and r_p to refer to the same quantity, here chosen to be the electric field.[7]) It is possible to construct a perturbation theory for r_p in terms of the solution $B_0 = \exp(iq_0z)$ for the case in which $v = v_0$ everywhere. The corresponding Green function (cf. the appendix of Ref. 9)

$$G_p(z, \zeta) = \frac{\exp(iq_0|z - \zeta|)}{2iQ_0}, \qquad Q_0 = q_0/\epsilon_0 \tag{32}$$

is a solution of

$$\frac{\partial}{\partial z}\left(v_0 \frac{\partial G}{\partial z}\right) + \left(\frac{\omega^2}{c^2} - v_0 K^2\right) G = \delta(z - \zeta). \tag{33}$$

B now satisfies the integrodifferential equation [Eq. (A.10) of Ref. 9]

$$B(z) = B_0(z) + \int_{-\infty}^{\infty} d\zeta \Delta v(\zeta) \left\{K^2 B(\zeta) G(z, \zeta) + \frac{dB}{d\zeta}\frac{\partial G}{\partial \zeta}\right\}. \tag{34}$$

An exact expression for r_p is obtained from Eq. (34) by extracting the coefficient of $\exp(-iq_0z)$ in the limit as $z \to -\infty$:

$$r_p = \frac{-1}{2iQ_0}\int_{-\infty}^{\infty} d\zeta \Delta v(\zeta) \left\{K^2 B B_0 + \frac{dB}{d\zeta}\frac{dB_0}{d\zeta}\right\}. \tag{35}$$

This may be written

$$r_p = \frac{1}{2iQ_0}\int_{-\infty}^{\infty} d\zeta \left\{\left(\frac{1}{\epsilon_0} - \frac{1}{\epsilon}\right) K^2 B B_0 + (\epsilon - \epsilon_0) C C_0\right\}, \tag{36}$$

where $C = (1/\epsilon)dB/dz$, $C_0 = (1/\epsilon_0)dB_0/dz$, in which form we see the equivalence to the comparison identity expression (46) of Ref. 9. We obtain the first-order perturbation theory expression for r_p by replacing B by B_0 and C by C_0 in Eq. (36). (This is equivalent to lowest order in Δv to replacing $dB/d\xi$ by $dB_0/d\xi$ but is preferable since C is continuous at a discontinuity in the dielectric function, whereas $dB/d\xi$ is not. A direct consequence is that our r_p^{pert} gives the correct first term in the film thickness–wavelength expansion,[8] to all orders in Δv.) We obtain

$$r_p^{\text{pert}} = \frac{1}{2iQ_0}\int_{-\infty}^{\infty} d\zeta \left\{\left(\frac{1}{\epsilon_0} - \frac{1}{\epsilon}\right) K^2 - (\epsilon - \epsilon_0 Q_0^2)\right\} \exp(2iq_0\zeta). \tag{37}$$

To derive a variational expression for r_p, we rewrite the integrodifferential equation (34) with the unknown B on the left side and operate with

$$\int_{-\infty}^{\infty} dz \left\{\Delta v(z) K^2 B(z) - \frac{d}{dz}\left(\Delta v(z)\frac{dB}{dz}\right)\right\} \tag{38}$$

on both sides. We again write the resulting equation as $S = F$, where S (the left-hand side) is second degree in B, and F (the right-hand side) is first degree in B. We have, after an integration by parts,

$$F = \int_{-\infty}^{\infty} dz \Delta v \left\{K^2 B B_0 + \frac{dB}{dz}\frac{dB_0}{dz}\right\} = 2iQ_0 r_p \tag{39}$$

[by Eq. (35)]. The second-degree term S becomes, again after integration by parts,

$$S = \int_{-\infty}^{\infty} dz \Delta v K^2 B \left\{B - \int_{-\infty}^{\infty} d\zeta \Delta v \left[K^2 B G + \frac{dB}{d\zeta}\frac{\partial G}{\partial \zeta}\right]\right\}$$
$$+ \int_{-\infty}^{\infty} dz \Delta v \frac{dB}{dz}\left\{\frac{dB}{dz} - \int_{-\infty}^{\infty} d\zeta \Delta v \left[K^2 B \frac{\partial G}{\partial z} + \frac{dB}{d\zeta}\frac{\partial^2 G}{\partial z \partial \zeta}\right]\right\}. \tag{40}$$

A calculation along similar lines to that for the s wave (but more complex) establishes that $\delta S = 2\delta F$. Thus $\delta(F^2/S) = 0$, and the variational expression for r_p is, on using Eq. (39),

$$r_p^{\text{var}} = -\frac{F^2/S}{2iQ_0}. \tag{41}$$

At normal incidence ($K = 0$), $i\sqrt{\epsilon_0}(\omega/c)B$ is equal to dE/dz, where E is the solution of $d^2E/dz^2 + \epsilon(\omega^2/c^2)E = 0$ (see Sec. 3 of Ref. 7). Using this and the fact that

$$\frac{\partial^2 G_p}{\partial z \partial \zeta} = q_0^2 G_p - \epsilon_0 \delta(z - \zeta), \tag{42}$$

we find that $F_p = F_s/\epsilon_0$, $S_p = S_s/\epsilon_0$. Thus r_p and r_s are identical at normal incidence, as are r_p^{var} and r_s^{var} for trial functions satisfying the above relations.

The simplest variation trial function for $B(z)$ is $B_0(z) = \exp(iq_0z)$. This gives the values F_0 and S_0 for F and S, where, using $\epsilon\epsilon_0\Delta v = \epsilon_0 - \epsilon = -\Delta\epsilon$,

$$F_0 = -2iQ_0 r_p^{\text{pert}} = \int_{-\infty}^{\infty} dz(\Delta v K^2 + \Delta\epsilon Q_0^2)\exp(2iq_0z) \tag{43}$$

[see Eq. (37) and the discussion preceding it]. To evaluate S_0 we rewrite S [using Eq. (42)] as

$$S = \int_{-\infty}^{\infty} dz\{\Delta v K^2 B^2 - \Delta\epsilon C^2\}$$
$$+ 2K^2 \int_{-\infty}^{\infty} dz \Delta v B \int_{-\infty}^{\infty} d\zeta \Delta\epsilon C \frac{1}{\epsilon_0}\frac{\partial G}{\partial \zeta}$$
$$- Q_0^2 \int_{-\infty}^{\infty} dz \Delta\epsilon C \int_{-\infty}^{\infty} d\zeta \Delta\epsilon C G$$
$$- K^4 \int_{-\infty}^{\infty} dz \Delta v B \int_{-\infty}^{\infty} d\zeta \Delta v B G. \tag{44}$$

We now replace B by $B_0 = \exp(iq_0z)$ and C by $C_0 = iQ_0 \exp(iq_0z)$ to obtain

$$S_0 = F_0 + \frac{2iK^2Q_0}{\epsilon_0}\int_{-\infty}^{\infty} dz \Delta v \exp(iq_0z)$$
$$\times \int_{-\infty}^{\infty} d\zeta \Delta\epsilon \exp(iq_0\zeta)\partial G/\partial\zeta$$
$$- K^4 \int_{-\infty}^{\infty} dz \Delta v \exp(iq_0z) \int_{-\infty}^{\infty} d\zeta \Delta v \exp(iq_0\zeta) G$$
$$+ Q_0^4 \int_{-\infty}^{\infty} dz \Delta\epsilon \exp(iq_0z) \int_{-\infty}^{\infty} d\zeta \Delta\epsilon \exp(iq_0\zeta) G. \tag{45}$$

The corresponding variational estimate for the reflection amplitude is

$$r_p^{\text{var}} = \frac{F_0}{S_0} r_p^{\text{pert}}. \tag{46}$$

At grazing incidence ($Q_0 \to 0$) this variational expression tends to $+1$, in accord with the general result[7] that the reflected electric and magnetic fields are then precisely out of phase with the incident fields.

In the long-wave limit, the leading terms of F_0 and S_0 are, to second order in the film thickness,

$$F_0 = \epsilon_0^{-2}\{q_0^2\lambda_1 - K^2\Lambda_1\} + 2iq_0\epsilon_0^{-2}\{q_0^2\lambda_2 - K^2\Lambda_2\} + \ldots, \quad (47)$$

$$S_0 - F_0 = (2iq_0\epsilon_0^3)^{-1}(q_0^4\lambda_1^2 - K^4\Lambda_1^2 - 2(\epsilon_0 q_0 K)^2 J_2) + \ldots, \quad (48)$$

where (cf. Ref. 8, appendixes A and B)

$$\lambda_n = \int_{-\infty}^{\infty} dz(\epsilon - \epsilon_0)z^{n-1}, \quad \Lambda_n = \epsilon_0^2 \int_{-\infty}^{\infty} dz \left(\frac{1}{\epsilon_0} - \frac{1}{\epsilon}\right) z^{n-1} \quad (49)$$

and

$$J_2 = \int_{-\infty}^{\infty} dz \int_{-\infty}^{\infty} d\zeta \,\mathrm{sgn}(z - \zeta) \left[\frac{1}{\epsilon_0} - \frac{1}{\epsilon(z)}\right][\epsilon(\zeta) - \epsilon_0]. \quad (50)$$

From these results we find that the variational reflection amplitude for the p wave as given by Eq. (46) is correct to second order in the film thickness [Ref. 8, Eqs. (41) and (43)].

4. REFLECTION AMPLITUDES IN TERMS OF FIVE INTEGRALS

The first-order perturbation theory approximations for the reflection amplitudes can be expressed in terms of two Fourier integrals (cf. Ref. 8, appendix A)

$$\lambda(k) = \int_{-\infty}^{\infty} dz \exp(ikz)\Delta\epsilon, \quad (51)$$

$$\Lambda(k) = \epsilon_0 \int_{-\infty}^{\infty} dz \exp(ikz) \frac{\Delta\epsilon}{\epsilon}. \quad (52)$$

These have the dimensions of length (or of length × dielectric constant, if the latter is given a dimensionality). In terms of λ and Λ,

$$r_s^{\mathrm{pert}} = -\frac{\omega^2/c^2}{2iq_0}\lambda(2q_0), \quad (53)$$

$$r_p^{\mathrm{pert}} = -\frac{1}{2iq_0\epsilon_0}[q_0^2\lambda(2q_0) - K^2\Lambda(2q_0)]. \quad (54)$$

The variational expressions based on the same wave function as the first-order perturbation theory require three more integrals. These are not Fourier transforms but have a related character:

$$\sigma(k) = \int_{-\infty}^{\infty} dz\Delta\epsilon \left\{\exp(ikz)\int_{-\infty}^{z} d\zeta\Delta\epsilon + \int_{z}^{\infty} d\zeta \exp(ik\zeta)\Delta\epsilon\right\}, \quad (55)$$

$$\Sigma(k) = \epsilon_0^2 \int_{-\infty}^{\infty} dz \frac{\Delta\epsilon}{\epsilon}\left\{\exp(ikz)\int_{-\infty}^{z} dz \frac{\Delta\epsilon}{\epsilon} + \int_{z}^{\infty} d\zeta \exp(ik\zeta)\frac{\Delta\epsilon}{\epsilon}\right\}, \quad (56)$$

$$\Gamma(k) = \epsilon_0 \int_{-\infty}^{\infty} d\zeta \frac{\Delta\epsilon}{\epsilon}\left\{\exp(ikz)\int_{-\infty}^{z} d\zeta\Delta\epsilon - \int_{z}^{\infty} d\zeta \exp(ik\zeta)\Delta\epsilon\right\}. \quad (57)$$

They all have the dimensions of (length × dielectric constants).[2] (In both the σ and Σ expressions, the first and second terms are equal because of the z, ζ symmetry of the integrands.) In terms of these integrals,

$$r_s^{\mathrm{var}} = \frac{-\dfrac{\omega^2/c^2}{2iq_0}\lambda(2q_0)}{1 + \dfrac{\omega^2/c^2}{2iq_0}\dfrac{\sigma(2q_0)}{\lambda(2q_0)}} \quad (58)$$

and

$$r_p^{\mathrm{var}} = \frac{-\dfrac{1}{2iq_0\epsilon_0}[q_0^2\lambda(2q_0) - K^2\Lambda(2q_0)]}{1 + \dfrac{[q_0^4\sigma(2q_0) - K^4\Sigma(2q_0) - 2q_0^2K^2\Gamma(2q_0)]}{2iq_0\epsilon_0[q_o^2\lambda(2q_0) - K^2\Lambda(2q_0)]}}. \quad (59)$$

(The numerators in each case give the first-order perturbation result.) At normal incidence, when $K \to 0$ and $q_0 \to \sqrt{\epsilon_0}\omega/c \equiv k_0$, both reflection amplitudes tend to

$$r_n^{\mathrm{var}} = \frac{-\dfrac{k_0}{2i\epsilon_0}\lambda(2k_0)}{1 + \dfrac{k_0}{2i\epsilon_0}\dfrac{\sigma(2k_0)}{\lambda(2k_0)}}. \quad (60)$$

At grazing incidence, when $q_0 \to 0$, the results $r_s^{\mathrm{var}} \to -1$ and $r_p^{\mathrm{var}} \to 1$ follow from

$$\sigma(0) = \lambda^2(0); \quad \Sigma(0) = \Lambda^2(0). \quad (61)$$

From Eq. (59) we see that a film between two like media is transparent to the p wave (according to both the first-order perturbation and the variational theories) at an angle

$$\theta = \arctan[\lambda(2q_o)/\Lambda(2q_0)]^{1/2}. \quad (62)$$

This is an approximate extension of the rigorous result[8] that, to lowest order in the film thickness, there is transparency at $\theta = \arctan(\lambda_1/\Lambda_1)^{1/2}$. Note, however, that the ratio λ/Λ is not (in general) real. Complete transparency at a certain angle is thus characteristic of thin films; as we shall see in the next section, it also characterizes uniform films of any thickness.

5. COMPARISON WITH EXACT RESULTS FOR UNIFORM FILM

For the important special case of a uniform film of constant dielectric function ϵ, located between z_1 and $z_2 = z_1 + \Delta z$ in a medium of dielectric function ϵ_0, we have[8]

$$r_s = \exp(2iq_0z_1)\frac{i(q^2 - q_0^2)\tau}{2qq_0 - i(q^2 + q_0^2)\tau},$$

$$-r_p = \exp(2iq_0z_1)\frac{i(Q^2 - Q_0^2)\tau}{2QQ_0 - i(Q^2 + Q_0^2)\tau}, \quad (63)$$

and

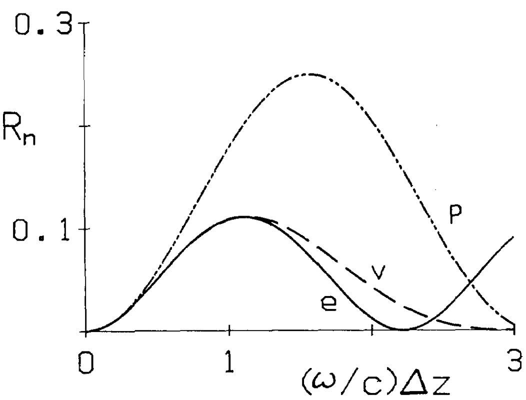

Fig. 1. Reflectivity at normal incidence as a function of the film thickness Δz. The exact reflectivity (e) is the solid curve, the perturbation result (p) is the dashed–dotted curve, and the variational result (v) is the dashed curve. In this and the following figures, $\epsilon_0 = 1$ and $\epsilon = 2$.

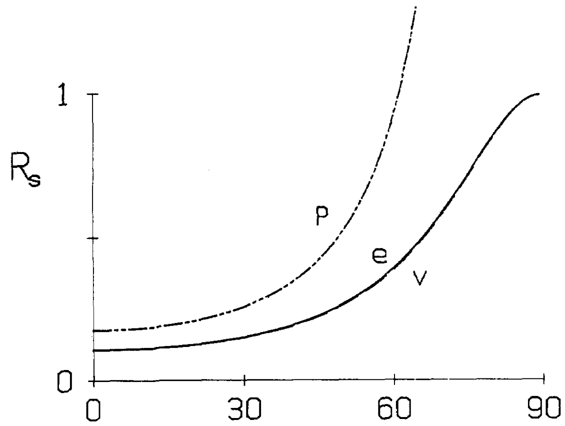

Fig. 2. Reflectivity for the s wave as a function of the angle of incidence at $(\omega/c)\Delta z = 1$. The exact, perturbation, and variational results are denoted by curves e, p, and v, respectively.

$$\frac{r_p}{r_s} = \left[\cos^2\theta - \frac{\epsilon_0}{\epsilon}\sin^2\theta\right]\left\{\frac{1-\frac{i}{2}\left(\frac{q}{q_0}+\frac{q_0}{q}\right)\tau}{1-\frac{i}{2}\left(\frac{Q}{Q_0}+\frac{Q_0}{Q}\right)\tau}\right\}, \quad (64)$$

where

$$q^2 = \epsilon\frac{\omega^2}{c^2} - K^2, \qquad Q = q/\epsilon, \qquad \tau = \tan(q\Delta z). \quad (65)$$

Note that r_p is zero at $\theta = \arctan\sqrt{\epsilon/\epsilon_0}$, which is the same as the Brewster angle for light incident from a medium with dielectric constant ϵ_0 onto a bulk medium of dielectric constant ϵ. A uniform film between like media is always transparent to the p wave at the same angle, irrespective of its thickness.

For the perturbation and variational expressions we need the five integrals defined in the last section. These take the values

$$\lambda(k) = \Delta\epsilon\exp(ikz_1)[\exp(ik\Delta z) - 1]/ik, \quad (66)$$

$$\sigma(k) = 2(\Delta\epsilon)^2\exp(ikz_1)\{\Delta z\exp(ik\Delta z) - [\exp(ik\Delta z) - 1]/ik\}/ik, \quad (67)$$

$$\Lambda(k) = \frac{\epsilon_0}{\epsilon}\lambda(k), \qquad \Sigma(k) = \left(\frac{\epsilon_0}{\epsilon}\right)^2\sigma(k), \qquad \Gamma(k) = 0. \quad (68)$$

From Eq. (59) we thus obtain for the p-wave reflection amplitude

$$r_p^{\text{var}} = \frac{-\frac{\omega/c}{2i\sqrt{\epsilon_0}\cos\theta}\left[\cos^2\theta - \frac{\epsilon_0}{\epsilon}\sin^2\theta\right]\lambda(2q_0)}{1+\frac{\omega/c}{2i\sqrt{\epsilon_0}\cos\theta}\left[\cos^2\theta + \frac{\epsilon_0}{\epsilon}\sin^2\theta\right]\frac{\sigma(2q_0)}{\lambda(2q_0)}}. \quad (69)$$

This correctly gives transparency at $\theta = \arctan(\epsilon/\epsilon_0)^{1/2}$. For the ratio of the amplitudes we find [compare with Eq. (64)]

$$\frac{r_p^{\text{var}}}{r_s^{\text{var}}} = \left[\cos^2\theta - \frac{\epsilon_0}{\epsilon}\sin^2\theta\right] \times \frac{1+\frac{\omega/c}{2i\sqrt{\epsilon_0}\cos\theta}\frac{\sigma(2q_0)}{\lambda(2q_0)}}{1+\frac{\omega/c}{2i\sqrt{\epsilon_0}\cos\theta}\left[\cos^2\theta + \frac{\epsilon_0}{\epsilon}\sin^2\theta\right]\frac{\sigma(2q_0)}{\lambda(2q_0)}}. \quad (70)$$

[The s-wave reflection amplitude is given directly by Eq. (58).] We see that in each case we need $\lambda(2q_0)$ and the ratio

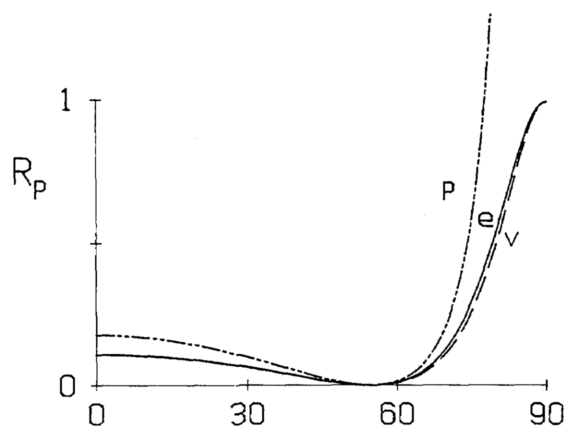

Fig. 3. Reflectivity for the p wave as a function of the angle of incidence, at $(\omega/c)\Delta z = 1$. The exact, perturbation, and variational reflectivities are all zero at $\theta = \arctan\sqrt{2} \simeq 54.7°$.

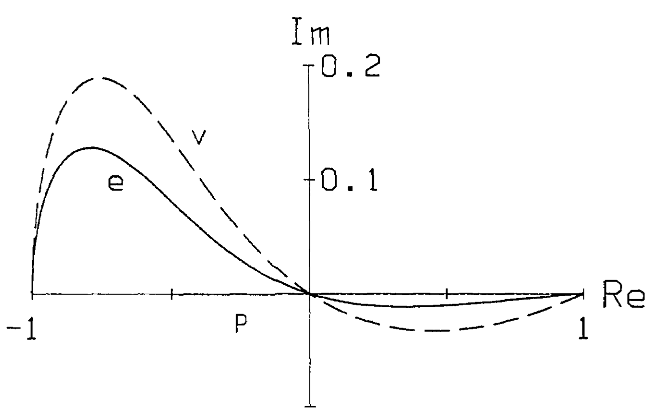

Fig. 4. The ratio r_p/r_s in the complex plane, at $(\omega/c)\Delta z = 1$. The exact (e) and variational (v) trajectories are shown by solid and dashed lines, respectively; the perturbation trajectory lies along the real axis between +1 and −½. All three trajectories start at +1 at normal incidence and pass through the origin at $\theta = \arctan(\epsilon/\epsilon_0)^{1/2} \simeq 54.7°$. Only the perturbation trajectory does not end at −1 at grazing incidence.

$\sigma(2q_0)/\lambda(2q_0)$. It is convenient to rewrite Eqs. (66) and (67) in the form

$$\lambda(2q_0) = \Delta\epsilon\Delta z \exp[iq_0(z_1 + z_2)]j_0(q_0\Delta z), \tag{71}$$

$$\sigma(2q_0) = (\Delta\epsilon\Delta z)^2 \exp[iq_0(z_1 + z_2)]\{j_0(q_0\Delta z) + ij_1(q_0\Delta z)\}, \tag{72}$$

where $j_0(x) = \sin x/x$ and $j_1(x) = \sin x/x^2 - \cos x/x$ are spherical Bessel functions. It is then clear that σ/λ, and hence $R_s = |r_s|^2$, $R_p = |r_p|^2$, and r_p/r_s, are all independent of the location of the film, as they must be.

In Figs. 1–4 we compare the exact (e), perturbation (p), and variation (v) expressions for the reflectivity at normal incidence as a function of film thickness (Fig. 1), R_s and R_p as a function of angle of incidence (Figs. 2 and 3), and r_p/r_s in the complex plane as a function of angle (Fig. 4). The comparison is for the values $\epsilon_0 = 1$, $\epsilon = 2$.

We see that the simplest trial function [the plane wave $\exp(iq_0z)$] gives variational results that are far better than the perturbation results when the film thickness is small compared with the wavelength but that both results fail for thick films. For example, the zeros in the reflectivity occur at $q\Delta z = n\pi$ ($n = 1, 2, \ldots$), whereas the trial function $\exp(iq_0z)$ produces zeros at $q_0\Delta z = n\pi$. It is clear that a theory that takes account of the variation in wave number with variation in the dielectric function is required for thicker films.

6. EFFECT OF ANISOTROPY

We will consider only systems with azimuthal symmetry (about the normal to the surface), that is, those that are characterized by two dielectric functions, $\epsilon_\parallel(z)$ and $\epsilon_\perp(z)$ (in our geometry, $\epsilon_x = \epsilon_y = \epsilon_\parallel$, $\epsilon_z = \epsilon_\perp$). We denote equations (used earlier) modified to take account of anisotropy by adding primes to their numbers. On eliminating B_x and B_z from Eqs. (1) and

$$\frac{\partial B_x}{\partial x} - \frac{\partial B_z}{\partial x} = -i\epsilon_\parallel \frac{\omega}{c} E_y, \tag{2'}$$

we find that

$$\frac{\partial^2 E_y}{\partial z^2} + \frac{\partial^2 E_y}{\partial x^2} + \epsilon_\parallel \frac{\omega^2}{c^2} E_y = 0, \tag{3'}$$

and separation of variables by the substitution [Eq. (4)] gives the equation

$$\frac{d^2E}{dz_2} + \left(\epsilon_\parallel \frac{\omega^2}{c^2} - K^2\right)E = 0. \tag{5'}$$

All equations for the s wave derived earlier are thus modified only by the replacement of $\epsilon(z)$ by $\epsilon_\parallel(z)$.

The p wave is more complicated, since it samples (for a general angle of incidence) both $\epsilon_\parallel$ and $\epsilon_\perp$. Equation (26) becomes[10]

$$\frac{\partial B_y}{\partial z^2} = i\epsilon_\parallel \frac{\omega}{c} E_x, \qquad \frac{\partial B_y}{\partial x} = -i\epsilon_\perp \frac{\omega}{c} E_z. \tag{26'}$$

Elimination of E_x and E_z from Eqs. (26′) and (27) gives

$$\frac{\partial}{\partial z}\left(\frac{1}{\epsilon_\parallel}\frac{\partial B_y}{\partial z}\right) + \frac{\partial}{\partial x}\left(\frac{1}{\epsilon_\perp}\frac{\partial B_y}{\partial x}\right) + \frac{\omega^2}{c^2} B_y = 0. \tag{28'}$$

Separation of variables by the substitution [Eq. (29)] then gives

$$\frac{d}{dz}\left(v_\parallel \frac{dB}{dz}\right) + \left(\frac{\omega^2}{c^2} - v_\perp K^2\right) B = 0, \tag{30'}$$

where $v_\parallel = 1/\epsilon_\parallel$ and $v_\perp = 1/\epsilon_\perp$. The integrodifferential equation now satisfied by B is

$$B(z) = B_0(z) + \int_{-\infty}^{\infty} dzG(z, \zeta)\left[\Delta v_\perp K^2 B(\zeta) - \frac{d}{d\zeta}\left(\Delta v_\parallel \frac{dB}{d\zeta}\right)\right]$$
$$= B_0(z) + \int_{-\infty}^{\infty} d\zeta\left[\Delta v_\perp K^2 BG + \Delta v_\parallel \frac{dB}{d\zeta}\frac{\partial G}{\partial \zeta}\right], \tag{34'}$$

as may be verified by writing Eq. (30′) in the form

$$\frac{d}{dz}\left(v_0 \frac{dB}{dz}\right) + \left(\frac{\omega^2}{c^2} - v_0 K^2\right) B = \Delta v_\perp K^2 B - \frac{d}{dz}\left(\Delta v_\parallel \frac{dB}{dz}\right).$$

The exact r_p is [cf. Eq. (A12) of Ref. 10]

$$r_p = \frac{1}{2iQ_0}\int_{-\infty}^{\infty} d\zeta\left\{\left(\frac{1}{\epsilon_0} - \frac{1}{\epsilon_\perp}\right)K^2 BB_0 + (\epsilon_\parallel - \epsilon_0)CC_0\right\}, \tag{36'}$$

where now $C(\zeta) = (1/\epsilon_\parallel)dB/d\zeta$. The corresponding first-order perturbation expression is

$$r_p^{\text{pert}} = \frac{1}{2iQ_0}\int_{-\infty}^{\infty} d\zeta\left\{\left(\frac{1}{\epsilon_0} - \frac{1}{\epsilon_\perp}\right)K^2 - (\epsilon_\parallel - \epsilon_0)Q_0^2\right\}\exp(2iq_0\zeta). \tag{37'}$$

The variational expression is obtained by operating on the integrodifferential equation with

$$\int_{-\infty}^{\infty} dz\left\{\Delta v_\perp(z)K^2 B(z) - \frac{d}{dz}(\Delta v_\parallel(z)dB/dz)\right\}. \tag{38'}$$

The term that is first degree in B is again $F = -2iQ_0r_p$. The second-degree term becomes

$$S = \int_{-\infty}^{\infty} dz\Delta v_\perp K^2 B\left\{B - \int_{-\infty}^{\infty} d\zeta\left[\Delta v_\perp K^2 BG + \Delta v_\parallel \frac{dB}{d\zeta}\frac{\partial G}{\partial\zeta}\right]\right\} + \int_{-\infty}^{\infty} dz\Delta\psi_\parallel \frac{dB}{dz}\left\{\frac{dB}{dz} - \int_{-\infty}^{\infty} d\zeta\left[\Delta v_\perp K^2 B\frac{\partial G}{\partial z} + \Delta v_\parallel \frac{dB}{d\zeta}\frac{\partial^2 G}{\partial z\partial\zeta}\right]\right\}. \tag{40'}$$

We again have $\delta S = 2\delta F$ and $r_p^{\text{var}} = -F^2/2iQ_0S$. For the simplest trial function, $B_0(z) = \exp(iq_0z)$, we have

$$F_0 = \epsilon_0^{-2}[q_0^2\lambda(2q_0) - K^2\Lambda(2q_0)], \tag{73}$$

$$S_0 = F_0 + (2iq_0\epsilon_0^3)^{-1}[q_0^4\sigma(2q_0) - K^4\,\Sigma\,(2q_0) - 2K^2q_0^2\Gamma(2q_0)], \tag{74}$$

where now

$$\lambda(k) = \int_{-\infty}^{\infty} dz \exp(ikz)\Delta\epsilon_\parallel, \tag{51'}$$

$$\Lambda(k) = \epsilon_0\int_{-\infty}^{\infty} dz \exp(ikz)\frac{\Delta\epsilon_\perp}{\epsilon_\perp}, \tag{52'}$$

$$\sigma(k) = \int_{-\infty}^{\infty} dz\Delta\epsilon_{\|} \left\{\exp(ikz) \int_{-\infty}^{z} d\zeta\Delta\epsilon_{\|} + \int_{z}^{\infty} d\zeta \exp(ik\zeta)\Delta\epsilon_{\|}\right\}, \quad (55')$$

$$\Sigma(k) = \epsilon_0^2 \int_{-\infty}^{\infty} dz \frac{\Delta\epsilon_{\perp}}{\epsilon_{\perp}} \left\{\exp(ikz) \int_{-\infty}^{z} d\zeta \frac{\Delta\epsilon_{\perp}}{\epsilon_{\perp}} + \int_{z}^{\infty} d\zeta \exp(ik\zeta) \frac{\Delta\epsilon_{\perp}}{\epsilon_{\perp}}\right\}, \quad (56')$$

$$\Gamma(k) = \epsilon_0 \int_{-\infty}^{\infty} dz \frac{\Delta\epsilon_{\perp}}{\epsilon_{\perp}} \left\{\exp(ikz) \int_{-\infty}^{z} d\zeta\Delta\epsilon_{\|} - \int_{z}^{\infty} d\zeta \exp(ik\zeta)\Delta\epsilon_{\|}\right\}. \quad (57')$$

The perturbation and variational expressions for r_s and r_p have the same form as before [given by Eqs. (58) and (59)], with the integrals for the anisotropic case defined above. At normal incidence we have equality of the reflection amplitudes, and, as before, $r_s^{\text{var}} \to -1$ and $r_p^{\text{var}} \to 1$ at grazing incidence. There is again transparency at the angle given by Eq. (62).

For the uniform but anisotropic film, Eq. (63) remains valid, with $q_s^2 = \epsilon_{\|}\omega^2/c^2 - K^2$, $q_p^2 = \epsilon_{\|}\omega^2/c^2 - K^2\epsilon_{\|}/\epsilon_{\perp}$, and $Q^2 = (1/\epsilon_{\|})\omega^2/c^2 - K^2/\epsilon_{\|}\epsilon_{\perp}$. The film is transparent to the p wave when $Q^2 = Q_0^2[=(1/\epsilon_0)\omega^2/c^2 - K^2/\epsilon_0^2]$. This is at the angle

$$\theta = \arctan\left[\frac{\epsilon_{\perp}}{\epsilon_0}\left(\frac{\epsilon_{\|} - \epsilon_0}{\epsilon_{\perp} - \epsilon_0}\right)\right]^{1/2}. \quad (75)$$

The relationship between λ and Λ is now

$$\Lambda(k) = \frac{\Delta\epsilon_{\perp}}{\epsilon_{\perp}} \frac{\epsilon_0}{\Delta\epsilon_{\|}} \lambda(k), \quad (76)$$

and so the perturbation and variational expressions for r_p again give the correct angle for transparency. Note, however, that these expressions give interference zeros at $q_0\Delta z = n\pi$, whereas the s- and p-wave zeros occur at $q_s\Delta z = n\pi$ and $q_p\Delta z = n\pi$, respectively.

7. CONCLUSION

We have shown that the simplest variational expressions for the s and p reflection amplitudes are a substantial improvement over those of first-order perturbation theory. In particular, the troublesome divergence of the perturbation amplitudes at grazing incidence is replaced by correct limiting values. The expressions derived include the possibility of absorption and/or anisotropy within the film. All information is in terms of five integrals over the (arbitrary) dielectric function profile. The variational expressions are correct to second order in film thickness/wavelength and work well for films that are thin compared with the wavelength. For thicker films, it may be possible to construct variational expressions using Green functions appropriate to the shortwave limit.

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