Reflection of light at a flat interface under normal incidence: A renewed macroscopic description

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We consider a cubic crystal, layered orthogonally to the incident wave vector, with some arbitrary refractive-index profile. The local electric field experienced by a crystal molecule may be calculated in the macroscopic limit, using a set of linear equations. To evaluate the amplitude reflection coefficient R, we use first- and higher-order Born approximations, and then show that R is the sum $(R_1 + R_3 + \cdots)$ of coefficients related to paths presenting one extremum, three extrema, etc. This approach gives a new physical insight into the reflection of light in terms of scattering and nothing but scattering. It also leads to an improved evaluation of the interfacial reflectivity. The different approximate formulas that are derived are compared numerically for a dissymmetric refraction-index profile.

I. INTRODUCTION

The reflection of light by plane interfaces has been used extensively to gain the structural information needed for the optical description of these interfaces.^{1,2} Three main techniques have been developed: ellipsometry, reflectometry, $^{3-12}$ and surface-plasmon oscillations. 13,14 To analyze the experimental results obtained using these techniques, namely the reflection coefficients, several theories are available: $^{15-19}$ they proceed by integration of the macroscopic Maxwell equations. In a recent paper,²⁰ we proposed a self-consistent method to determine the reflection coefficient of a flat interface by solving the microscopic Maxwell equations. This approach involves the resolution of a set of linear equations. In the present paper we determine the reflection coefficients, in the macroscopic limit, by solving this set of equations with the Born approximation method.²¹ This analysis provides a better insight into the mechanism of light reflection: It shows in terms of scattering, and nothing but scattering, how light is reflected from a medium of variable refractive index. It also leads to new approximate formulas for the evaluation of the reflection coefficients with the physical explanation of these approximations. We restrict our present analysis to the case of normal incidence. In a forthcoming paper, this approach is planned to be extended to any incidence angle, for both s and p waves.

II. GENERAL EQUATIONS FROM THE SELF-CONSISTENT EQUATION TO THE PATH DESCRIPTION

Let us consider a plane interface between vacuum and a crystal of molecules positioned on a cubic lattice of characteristic lattice parameter a. The polarizabilities of the molecules, $\alpha_m(l)$, are only functions of the crystal plane location number l ($l \in \mathbf{N}$, see Fig. 1). In other words, we consider a layered crystal with some arbitrary refractive-index profile. The set of self-consistent equations allowing the calculation of the local electric field $\mathbf{E}(l)$, applied to any molecule of the crystal plane l, may be written [Eq. (15) of Ref. 20]

$$-\exp(jpk_0a) = \sum_{l=0}^{\infty} S_{p-l}(l)\beta(l)$$
(1)

with $k_0 = 2\pi/\lambda + j\eta$, λ being the wavelength of light in vacuum; η is supposed to be positive and represents a small absorption of the material; $\beta(l)$ is defined by

$$\alpha_m(l)\mathbf{E}(l)/4\pi a^3 = \beta(l)\mathbf{E}_i^0$$
,



FIG. 1. Schematic representation of a cubic lattice and its crystalline (X, Y) planes layered orthogonally to the incident wave vector \mathbf{k}_0 ; *a* is the fundamental repeat period and \mathbf{E}_i the incident electric field.

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(6)

where \mathbf{E}_i^0 is the incident electric field at the position l = z = 0. For $p \neq l$, the parameter $S_{p-l}(l)$ characterizes the interactions between one molecule within the plane p and all the molecules of the plane l. In the macroscopic approximation, its value is given by²⁰

$$S_{p-l}(l) = S_{p-l} = 2\pi j a k_0 \exp(j a k_0 |p-l|) \text{ for } p \neq l ,$$

$$S_0(l) = -\{4\pi / [n^2(l) - 1]\} + 2\pi j a k_0 ,$$
(2)

n(l) being the macroscopic refractive index (Lorentz in-

dex) of the plane
$$l$$
. The system (1) can be rewritten in the form

$$\beta(p) = [-S_0(p)]^{-1} \left[\exp(jk_0ap) + \sum_{l \neq p} S_{p-l}\beta(l) \right],$$

$$\forall p \in \mathbf{N} . \quad (3)$$

We can solve this set of linear equations using a Born method. We then obtain for the successive degrees of approximation

$$\begin{split} \beta^{(1)}(p) &= [-S_0(p)]^{-1} \exp(jk_0 ap) ,\\ \beta^{(2)}(p) &= [-S_0(p)]^{-1} \exp(jk_0 ap) + \sum_{l \neq p} [-S_0(p)]^{-1} S_{p-l} [-S_0(l)]^{-1} \exp(jk_0 al) ,\\ \beta^{(3)}(p) &= [-S_0(p)]^{-1} \exp(jk_0 ap) \\ &+ \sum_{l \neq p} [-S_0(p)]^{-1} (S_{p-l}) [-S_0(l)]^{-1} \exp(jk_0 al) \\ &+ \sum_{l \neq p} \sum_{m \neq l} [-S_0(p)]^{-1} (S_{p-l}) [-S_0(l)]^{-1} (S_{l-m}) [-S_0(m)]^{-1} \exp(jk_0 am) . \end{split}$$

$$(4)$$

The parameter $\beta^{(1)}(p)$ represents a solution of the set of equations (3) in the first Born approximation. It is proportional to the dipolar moment of a molecule in the *p* plane, induced by the incident electric field and by the field of all other dipoles within the plane *p*. In the same way, it can be shown that $\beta^{(2)}(p)$, which is the solution of Eq. (3) in the second Born approximation, represents the sum of $\beta^{(1)}(p)$ and of parameters proportional to the dipolar moment induced in a molecule of the *p* plane by all the dipoles located within the plane *l* and having a dipolar moment proportional to $\beta^{(1)}(l)$, etc.

From Eqs. (4), we can determine the amplitude reflection coefficient R using Eq. (19) of Ref. 20. This leads to

$$R = \exp(jk_0a) \left[\sum_{p=0}^{\infty} \exp(jk_0ap)(S_{-p})[-S_0(p)]^{-1} + \sum_{p=0}^{\infty} \sum_{l \neq p} (S_{-p})[-S_0(p)]^{-1}(S_{p-l})[-S_0(l)]^{-1}\exp(jk_0al) + \sum_{p=0}^{\infty} \sum_{l \neq p} \sum_{m \neq l} (S_{-p})[-S_0(p)]^{-1}(S_{p-l})[-S_0(l)]^{-1}(S_{l-m})[-S_0(m)]^{-1}\exp(jk_0am) + \cdots \right].$$
(5)

Let us now introduce the parameters $D^{(l)}$ defined by

$$\exp(jak_0|p-l|)D^{(l)} = (S_{p-l})[-S_0(l)]^{-1}.$$

Relation (2) shows that $D^{(1)}$ is indeed independent of p. The amplitude reflection coefficients R may be rewritten

$$\exp(-jk_{0}a)R = \sum_{p=0}^{\infty} \exp(2jk_{0}p)D^{(p)} + \sum_{p=0}^{\infty} \sum_{l\neq p} \exp[jk_{0}a(p+|p-l|+l)]D^{(p)}D^{(l)} + \sum_{p=0}^{\infty} \sum_{l\neq p} \sum_{m\neq l} \exp[jk_{0}a(p+|p-l|+|l-m|+m)]D^{(p)}D^{(l)}D^{(m)} + \cdots$$
(7)

It is important to observe that $D^{(m)}$ is proportional to the electrical field resulting from the diffusion by the plane m in the z > 0 or z < 0 directions. The length a(p+|p-l|+|l-m|+m) is equal to the length La of the path (p,l,m). Consequently, relation (7) is interpreted

as a sum over different paths starting from—and coming back to—the plane z = 0, and takes the closed form:

$$\exp(-jk_0a)R = \sum_{\substack{\text{all} \\ \text{paths}}} \exp(jk_0La) \prod_p D^{(p)} , \qquad (8)$$

where La is the length of the path and where p represents the planes along the path on which diffusion occurs. In the case of normal incidence, the value of $D^{(p)}$ is simply

$$D^{(p)} = \frac{2\pi j a k_0}{4\pi / [n^2(p) - 1] - 2\pi j a k_0} , \qquad (9)$$

where n(p) represents the macroscopic refractive index in the crystal plane p. We are here mostly interested in the limit $a \rightarrow 0$. Then, Eq. (9) leads to

$$D^{(p)} \simeq jak_0[n^2(p)-1]/2$$
 (10)

The calculation of the reflection coefficient as a sum, over all the paths, of the quantity $\exp(jk_0La)\prod_p D^{(p)}$ can be conducted as follows.

We first calculate the contribution R_1 to the amplitude reflectivity of all the paths with only one extremum. This is illustrated by Fig. 2. The number of such paths of length 2La is 2^{2L} . Each path can be represented by a set of numbers. For example, the contribution of the path (p,q,n,L,p) will be

$$\exp(jk_0La)D^{(p)}D^{(q)}D^{(n)}D^{(L)}D^{(p)}$$

. .

 $F(p) = \sum_{k=0}^{p} \exp(jk_0 la)$

Only the plane L must be taken into account for each path of length 2La. Therefore, R_1 is given by

$$\exp(-jk_0a)R_1 = D^{(0)} + \sum_{L=1}^{\infty} \exp(2jak_0L)D^{(L)}\prod_{i=0}^{L-1} (1+D^{(i)})^2,$$
(11)

where the $(1+D^{(i)})$ term arises from the fact that each plane can or cannot be taken into account. This provides a more general and better approximation than the second Born approximation, because it takes into account backscattering of higher order than only the second one.¹⁸ This means that R_1 contains the second Born approxima-



FIG. 2. Schematic representation of geometric paths involving one extremum and three extrema, the interface being located at z = -a/2. (a) Corresponds to a path of length zero, with one extremum, and thus to a backscattering towards the z < 0direction by the plane located at z = 0. (b) Presents a path with one extremum of length 2La (L = 6 in the figure). (c) Illustrates a path with three extrema of total length 2(p+m)a (in the figure we chose p = 4, l = 2, m = 2). Note that symbols \times , \bigcirc , and \downarrow introduce factors $D^{(k)}$, $1 + D^{(k)}$, and $\exp(jk_0a)$, respectively, in the expression of the reflection coefficient $R [D^{(k)} \text{ corre-}$ sponds to a plane numbered k and it is defined by Eq. (6)].

tion and partly higher-order Born approximation terms.

The contribution of the paths with two extrema is zero because all the paths must begin and end at the plane p = 0. Thus, the next contribution to the reflectivity, R_3 , comes from the paths displaying three extrema (see Fig. 2). Its value can be obtained by the method just developed for R_1 :

$$\exp(-jk_0a)R_3 = \sum_{p=1}^{\infty} \exp(jk_0pa) \left[\prod_{i=p-l}^{0} (1+D^{(i)}) \right] F(p)$$
(12)

with

$$\sum_{j=p-l+1}^{l=1} \times \prod_{j=p-l+1}^{p-l} (1+D^{(j)}) \times \sum_{m=l}^{\infty} \exp(jk_0ma) \prod_{k=p-l+m-1}^{p-l+1} (1+D^{(k)})\exp[jak_0(p-l+m)] \times \sum_{m=0}^{p-l+m-1} (1+D^{(n)})D^{(p)}D^{(p-l)}D^{(p-l+m)}.$$
(12)

III. EXAMPLES

A. Fresnel interface

Let us apply these results to a Fresnel interface between vacuum and a medium of refractive index n, in order to test how fast the development of R proposed converges: we obtain from Eqs. (10) and (11) the relation

$$R_{1F} = -\frac{n^2 - 1}{2(n^2 + 1)} . \tag{13}$$

For n = 1.6, the relative difference between R_{1F} and the Fresnel value of the amplitude reflection coefficient, $R_F = -(n-1)/(n+1)$, is about 4.5%; the closer n is to 1, the closer R_{1F} is to R_F . For this special example of an interface, we can also calculate R_{3F} . From Eq. (12), we derive.

(14)

 $\exp(-jk_0a)R_{3F}$ = $\sum_{p=1}^{\infty}\sum_{l=1}^{p}\sum_{m=1}^{\infty}\frac{D^3}{(1+D)^2}[(1+D)^{2l}\exp(2jk_0a)]^{p+m}$,

where $D = D^{(1)} = D^{(2)} = \cdots$. With $Q = \exp(2jk_0a) \times (1+D)^2$, Eq. (14) leads to

$$\exp(-jk_0a)R_{3F} = \frac{D^3}{(1+D)^2} \frac{Q}{1-Q} \sum_{p=1}^{\infty} pQ^p .$$
 (15)

Using the identity

$$\sum_{p=1}^{\infty} pQ^{p} = Q \frac{\delta}{\delta Q} \left[\frac{Q}{1-Q} \right], \qquad (16)$$

we finally obtain

$$\exp(-jk_0a)R_{3F} = \frac{D^3}{(1+D)^2} \frac{Q^2}{(1-Q)^3} .$$
 (17)

In the limit $a \rightarrow 0$, R_{3F} can then become

$$R_{3F} = -\frac{1}{8} \left[\frac{n^2 - 1}{n^2 + 1} \right]^3.$$
(18)

For n = 1.6, the difference between $(R_{1F} + R_{3F})$ and the real value R_F of the amplitude reflection coefficient is less than 0.5%. Thus, $(R_{1F} + R_{3F})$ provides already a very good approximation for the reflection coefficient in this particular case.

It may be checked that the total reflection coefficient $\sum R_{iF}$ has the following development:

$$\sum_{i} R_{iF} = -\frac{1/2}{1} \chi + \frac{(\frac{1}{2})(-\frac{1}{2})}{1 \times 2} \chi^{3} - \frac{(\frac{1}{2})(-\frac{1}{2})(-\frac{3}{2})}{1 \times 2 \times 3} \chi^{5} + \cdots$$
(19)

with

$$\chi = \frac{n^2 - 1}{n^2 + 1} \ . \tag{20}$$

Since $|\chi| < 1$, we have

$$\sum_{i} R_{iF} = -\frac{1}{\chi} + \frac{1}{\chi} (1 - \chi^2)^{1/2} = -\frac{n-1}{n+1} , \qquad (21)$$

a result which is indeed the Fresnel formula.²¹

B. Refractive-index profile

Let us now apply this approach to interfaces having index profiles which are different from the Fresnel type. Then, if we define φ by

$$D^{(i)} = ja\varphi(ia) \tag{22}$$

we have, in the limit $a \rightarrow 0$,

$$1 + D^{(i)} \simeq \exp[ja\varphi(ia)] . \tag{23}$$

Equation (11) leads to

$$\boldsymbol{R}_{1} = \sum_{L=0}^{\infty} \exp(2jk_{0}aL)aj\varphi(La)\exp\left[2j\sum_{i=0}^{L-1}a\varphi(ia)\right].$$
(24)

This can be rewritten in an integral form:

$$R_1 = \int_0^\infty j\varphi(z) \exp(2jk_0 z) \exp\left[2j\int_0^z \varphi(x)dx\right] dz \quad (25)$$

with

$$\varphi(z) = k_0 [n^2(z) - 1]/2 . \qquad (26)$$

In the same manner, we can demonstrate that in the limit $a \rightarrow 0$, we obtain for R_3

$$R_{3} = -\int_{0}^{\infty} dz_{1} \int_{0}^{z_{1}} dz_{2} \int_{z_{2}}^{\infty} dz_{3} j \varphi(z_{1}) \varphi(z_{2}) \varphi(z_{3}) \\ \times \exp\left[2j \int_{0}^{z_{1}} [k_{0} + \varphi(z)] dz - 2j \int_{0}^{z_{2}} [k_{0} + \varphi(z)] dz + 2j \int_{0}^{z_{3}} [k_{0} + \varphi(z)] dz\right].$$
(27)

It is more convenient to express the reflection coefficient by introducing the space derivative of the refractive index, dn/dz. Then, by an integration by parts, Eq. (25) leads to

$$R_{1} = -\int_{0}^{\infty} \frac{d}{dz} \left[\frac{\varphi(z)}{2[k_{0} + \varphi(z)]} \right] \exp\left[2j \left[k_{0}z + \int_{0}^{z} \varphi(x) dx \right] \right] dz + \left\{ \frac{\varphi(z)}{2[k_{0} + \varphi(z)]} \exp\left[2j \left[k_{0}z + \int_{0}^{z} \varphi(x) dx \right] \right] \right\}_{0}^{\infty}.$$
(28)

The last term in relation (28) is equal to zero, because the imaginary part η of the wave vector is positive. R_1 then becomes

$$R_{1} = -\int_{0}^{\infty} \frac{d}{dz} \left[\frac{\varphi(z)}{2[k_{0} + \varphi(z)]} \right] \exp\left[2j \left[k_{0}z + \int_{0}^{z} \varphi(x) dx \right] \right] dz \quad .$$
⁽²⁹⁾

From Eq. (13), it follows that

$$-\frac{\varphi(z)}{2[k_0+\varphi(z)]} = R_{1F}(z) , \qquad (30)$$

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where $R_{1F}(z)$ represents the coefficient R_1 for a Fresnel interface. From this relation we finally obtain

$$R_1 = \int_0^\infty \frac{dR_{1F}}{dn} \frac{dn}{dz} \exp\left[2j\left[k_0 z + \int_0^z \varphi(x) dx\right]\right] dz \quad .$$
(31)

In the same manner

$$R_{3} = 2 \int_{0}^{\infty} dz \left[\frac{d}{dz} R_{1F} \right] \exp\left[2j \int_{0}^{z} dx(\varphi + k_{0}) \right] \int_{0}^{z} dz_{1} j\varphi R_{1F} + \frac{4}{3} \int_{0}^{\infty} dz \left[\frac{d}{dz} R_{3F} \right] \exp\left[2j \int_{0}^{z} dx(\varphi + k_{0}) \right]$$
$$- \int_{0}^{\infty} dz \left[\frac{d}{dz} R_{1F} \right] \exp\left[2j \int_{0}^{z} dx(\varphi + k_{0}) \right] \int_{0}^{z} dz_{1} \left[\frac{d}{dz_{1}} R_{1F} \right] \exp\left[-2j \int_{0}^{z_{1}} dx(\varphi + k_{0}) \right]$$
$$\times \int_{z_{1}}^{\infty} dz_{2} \left[\frac{d}{dz_{2}} R_{1F} \right] \exp\left[2j \int_{0}^{z_{2}} dx(\varphi + k_{0}) \right]. \tag{32}$$

The foregoing formulas may also be applied for an interface between two media (0) and (1); k_0 then represents the wave vector in medium (0) and R_{1F} and φ are computed with the relative refractive index.

C. Approximations

Let us write the refractive-index profile into the form

$$n(z) = 1 + \Delta n f(z) , \qquad (33)$$

where Δn represents the refractive-index difference between the two bulk phases.

1. Approximation of the Webb type (Ref. 8)

Since the derivative of n(z) is

$$\frac{dn}{dz} = \Delta n f'(z) , \qquad (34)$$

the Webb approximation may be written as

$$\frac{dR_{1F}}{dn} = -\frac{1}{2} + \frac{\Delta n}{2} f(z) + O((\Delta n)^2)$$

leads to

$$R_{1} = -\frac{\Delta n}{2} \int_{0}^{\infty} f'(z) \exp\left[2j\left[k_{0}z + \int_{0}^{z} \varphi(x)dx\right]\right] dz + \frac{(\Delta n)^{2}}{4} \int_{0}^{\infty} (f^{2})'(z) \exp\left[2j\left[k_{0}z + \int_{0}^{z} \varphi(x)dx\right]\right] dz + O((\Delta n)^{3})$$
(40)

or, by introducing k(x),

$$R_{1} = -\frac{\Delta n}{2} \int_{0}^{\infty} f'(z) \exp\left[2j \int_{0}^{z} k(x) dx\right] dz + \frac{(\Delta n)^{2}}{4} \int_{0}^{\infty} (f^{2})'(z) \exp\left[2j \int_{0}^{z} k(x) dx\right] dz + O((\Delta n)^{3}).$$
(41)

Let us observe that $R_3 = O((\Delta n)^3)$.

3. Comparison between the foregoing approximations

The development of R_{1F} reads

$$R_{1F} = -\frac{\Delta n}{2} + \frac{(\Delta n)^2}{4} + O((\Delta n)^3) .$$
 (42)

The difference between the formulas giving R_1 in Eqs. (40) and (38) is then expressed by

$$\frac{dR_{1F}}{dn} \approx \frac{\Delta R_{1F}}{\Delta n} . \tag{35}$$

Thus, following Eq. (31),

$$R_1 \approx R_{1F} \int_0^\infty f'(z) \exp\left[2j\left[k_0 z + \int_0^z \varphi(x) dx\right]\right] dz \quad .$$
(36)

If we further develop the wave vector,

$$k_0 + \varphi(z) = k_0 n(z) + O((\Delta n)^2) , \qquad (37)$$

and write $k_0 n(x) = k(x)$, the reflection coefficient becomes

$$R_1 \approx R_{1F} \int_0^\infty f'(z) \exp\left[2j \int_0^z k(x) dx\right] dz \quad . \tag{38}$$

Let us observe that R_F and R_{1F} differ only by a thirdorder term in Δn .

2. Development of dR_{1F}/dn in powers of Δn

The equation

(39)

$$J = \frac{(\Delta n)^2}{2} \int_0^\infty f'(f - \frac{1}{2}) \\ \times \exp\left[2j\left[k_0 z + \int_0^z \varphi(x) dx\right]\right] dz \quad (43)$$

If the index profile is centrosymmetric

$$f(x) = -\frac{1}{2} + u(z - z_s) \text{ where } u \text{ is odd}$$

$$J = \frac{(\Delta n)^2}{2} \int_0^\infty (u^2)' \exp\left[2j\left[k_0 z + \int_0^z \varphi(x) dx\right]\right] dz .$$
(44)

It appears that in the case of a centrosymmetric refractive-index profile, J [then given by (44)] is practically negligible, and the Webb formula provides a very good approximation to the interfacial reflection. This is not the case for a nonsymmetric profile, unless Δn is very small, as shown earlier through numerical evaluations.²² Let us now illustrate this observation through a numerical example.

D. A numerical application

To check the validity of the approximation discussed above, we consider an example taken from ion implantation experiments. Starting with a plane Fresnel interface separating two homogeneous phases, one creates by implantation a significant refractive-index perturbation which is expected to relax exponentially from the interface into the bulk material. We thus study the following profile:

$$n(z) = 1, \quad z < 0$$

$$n(z) = n_2 + \Delta n \exp(-z/L), \quad z > 0.$$
(45)

The thickness L represents the first moment (mean distance to the interface) of the refractive-index distribution function. This is a highly nonsymmetric profile. Typically, when such a modified polymer material is put in contact with a solvent like water, the different refractive indexes are $n_1=1.33$ (solvent), $n_2=1.5$ (bulk polymer), and $\Delta n = 0.2$ (at the interface). The relative indexes are therefore 1, 1.12, and $\Delta n = 0.15$, respectively.

To characterize the reflection properties of the modified interface with respect to the initial one (before implantation) let us introduce dimensionless parameters Δ_i representing the relative difference between reflection coefficients (expressed as intensity ratios) calculated for the modified interface and for the original Fresnel interface. Keeping our previous notations, we define the Δ_i 's by

$$\Delta_i = (|R_i|^2 - R_F^2) / R_F^2, \quad i = A, W, 1, 1c .$$
(46)

 R_A is the exact amplitude reflection coefficient of the modified interface, calculated using the numerical Abelès method; ¹⁶ R_W corresponds to the same coefficient calculated with the Webb approximation; R_1 is evaluated by taking only the paths with one extremum into account [Eq. (41)], with an additional approximation for the phase integral to obtain simple analytical results in both cases

$$\int_{0}^{z} k(x) dx \simeq k_{0} (n_{2} + \Delta n/2) z \quad .$$
(47)

Finally, we define the parameter Δ_{1c} in which $|R_1|^2$ is replaced by $|R_{1c}|^2$ given by

$$|R_{1c}|^2 = |R_1|^2 + 2|R_1|^4 . (48)$$

This formula was derived for a Fresnel interface [Eq. (19)] and can be assumed to be a good approximation for interfaces with a thickness small compared to the wavelength. It takes rigorously into account all the paths with one extremum and provides an approximation for higher-order terms.

Figure 3 illustrates the variation of the different Δ ''s as a function of the interfacial thickness L. Several observations are in order.

(1) With the given refraction-index data, the approximative formulas display fairly good results for interfaces of thickness less than or of the order of 20 nm. Yet, as the thickness increases, Webb's approximate expression becomes progressively less satisfactory and, around L = 80 nm, it is even observed that $|R_W|$ exceeds the reflectivity for the Fresnel interface n_1/n_3 , a result which is obviously erroneous.

(2) If we now focus our attention on the values Δ_1 , Δ_{1c} , and Δ_A , it is seen that Δ_1 becomes significantly lower than Δ_A when L is larger than 50 nm, even if it is a much better approximation then Webb's formula. The two other estimates, Δ_{1c} and Δ_A , remain almost equal through the entire thickness range. The reason for the Δ_1 departure is clear: one has to account for all paths. This can partly be overcome by using the corrected reflection coefficient $|R_{1c}|^2$.

Hence, practical conclusions are the following. (i) If a nonsymmetrical refractive-index profile is anticipated, the Webb formula should not be used, as soon as the thickness becomes significant. (ii) The improved first-



FIG. 3. Representation of the variation of the relative reflectivity increments with respect to the unperturbed Fresnel interface for various thicknesses. —— corresponds to Webb's formula, —— corresponds to paths with one extremum only, —·—·· improves the latter with respect to higher-order terms, and —— is obtained from the numerical Abeles method and corresponds to the exact reflectivity. The horizontal line represents the Fresnel interface corresponding to infinite L.

order approximation we propose above, $(|R_{1c}|^2)$, is satisfactory; however, if $\Delta n/n_2$ becomes larger, higher-order terms have to be included explicitly. (iii) In a real experiment, $|R_F|^2$ and $|R_A|^2$ are directly measured. Nevertheless, since there are two unknowns for the profile we just studied (Δn and L), it is necessary to study reflection at different angles to determine with confidence the couple of values.

IV. CONCLUSION

In the present work, we calculated the reflectivity of a stratified interface as a sum of contributions related to diffusion paths at one dimension. Contributions of geometric paths presenting one, three, etc extrema were considered separately. When the refractive-index difference between both homogeneous media is small (typically if $\Delta n/n < 0.1$), it appears that the diffusion amplitude related to paths with one extremum provides a good description for the interfacial reflection. The improvement with respect to the Webb formula, developed for the case of centrosymmetric profiles where it appears to be well adapted, is discussed and ascertained in some detail. An explicit numerical calculation for a highly nonsymmetric profile shows the weakness of the Webb formula in such a situation, while our formula still works well, even in its simplest form. The present approach can be generalized to all incident angles, for both s and p polarizations; this will be shown in a future paper.

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