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ON REFLECTION FROM SURFACES WITH A THIN OVERLAYER

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Abstract

An approach to treat the reflectometry and ellipsometry data for bulk samples covered by an overlayer is suggested. The data can be used for the measurement of optical constants of solids, characterization of overlayers, and probe of the abruptness of spatial distribution of the bulk dielectric function. Numerical simulation shows that in the soft x-ray and XUV ranges the method can be applied for overlayers up to a thickness of 3–8 nm.

1. Introduction

Reflectometry and ellipsometry are the most convenient and widely used methods to determine the optical constants (i.e., real and imaginary parts of the dielectric function) of bulk materials. In reflectometry the intensity reflectivity $|R_p|^2$ (or $|R_s|^2$) of a sample is measured versus the angle of incidence or photon energy. In ellipsometry the quantity measured is the complex reflection ratio (or polarization factor)

$$\rho = \frac{R_s}{R_p} \tag{1}$$

where R_s and R_p are the amplitude reflection coefficients. A comprehensive survey of the methods with analysis of the accuracy within a wide spectral range can be found in [1–3]. To express the material dielectric function in terms of the reflection coefficients, each of the above methods essentially uses Fresnel formulas:

$$R_s = \frac{n_0 - n}{n_0 + n}, \quad R_p = \frac{\varepsilon n_0 - n}{\varepsilon n_0 + n}, \quad n_0 = \sin\theta, \quad n = \sqrt{\varepsilon - \cos^2\theta} \tag{2}$$

where θ is the grazing angle. The Fresnel formulas imply the two-phase model [3], where the mathematically sharp surface of a sample separates the bulk material from vacuum or ambience. However, due to various contamination or diffusion processes, real samples even after accurate preparation and cleaning cannot be considered as having stepwise spatial behavior with respect to chemical contents and material structure.

The effect of the overlayer (i.e., the transition zone located between the bulk material and vacuum) on the determination of the bulk optical constants has been extensively investigated (see [2–4] and references therein). One of the approaches to improve the accuracy consists in representing an overlayer as a homogeneous film with unknown thickness d and dielectric function $\varepsilon_{0\nu}$ which are found from processing

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the reflectivity data using the bulk optical constants [3–5]. However, various spectroscopic measurements show that the overlayer thickness d obtained in such a way depends on the wavelength in contrast to the model used. Microanalysis data also confirm the insufficient physical justification of the model. From the mathematical point of view the replacement of a gradually changing spatial profile of $\varepsilon(z)$ by a homogeneous layer with abrupt interfaces looks like an ambiguous procedure. The routes suggested to overcome the errors introduced by an overlayer are: (a) to concentrate on preparing high quality samples to eliminate the surface overlayer as far as possible and (b) to investigate independently the overlayer structure and introduce the data obtained into the electrodynamic model of a sample.

An alternative effort is made in this paper. We draw attention to some rigorous formulas and relations for reflectivity coefficients of bulks with an ideally sharp or slightly diffused surface. They are mainly based on general results [6] of scattering theory which have a long history but have still not been recognized for measurements within the VUV and soft x-ray regions. The reason for applying them is the small value of the overlayer thickness as compared with the radiation wavelength λ. It is important that to have the parameter a/λ not too small to provide observable corrections to the Fresnel formulas (2). This holds for various kinds of solid and liquid samples within the spectral range extending from far UV to soft x-rays. No assumptions are made in this approach on the spatial behavior of the dielectric function varying from the bulk value to vacuum. The formulas obtained for the reflection coefficients can be used in reflectometry and ellipsometry.

In Secs. 2 and 3, for the sake of completeness we give the full derivation of formulas for the reflection coefficients of s- and p-polarized light which are valid for thin transition layers $a/\lambda \ll 1$. Our approach does not use the Green function formalism (used in [6]) and can obviously be extended to more complicated cases, for instance, determination of optical constants in thin films.

The final formulas are summarized and discussed in Secs. 4 and 5.

A specific relation between the s-wave reflectivity R_s and complex reflection ratio ρ [see (1)] is established in Sec. 5. It is a consequence of the Fresnel reflection formulas and therefore can be used to advantage for the abruptness of an interface.

The results of simulation given in Sec. 6 for VUV and soft x-rays allow one to estimate the typical overlayer thickness for which the theory can be applied.

The main points of the present paper are briefly discussed in [7].

2. S-polarization

Let $\varepsilon(z)$ represent the spatial profile of the complex dielectric permittivity of a sample, tending to the constant value ε of a bulk material for $z \gg a$ and to the free space value $\varepsilon = 1$ for $z \ll a$. Then, for an incident beam with vector $\vec{E}$ perpendicular to the plane of incidence, the wave field $E(z)$, reflectivity R, and transmittivity T can be found as a solution of the 1D scattering problem:

$$E'' + k^2\left[\varepsilon(z) - \cos^2\theta\right]E = 0, \tag{3}$$

$$\begin{array}{lll} E \approx e^{ikn_0 z} + Re^{-ikn_0 z}, & n_0 = \sin\theta, & z \to -\infty, \\ E \approx Te^{iknz}, & n^2 = \varepsilon - \cos^2\theta, \quad \varepsilon = \varepsilon(z \to \infty), & z \to \infty. \end{array} \tag{4}$$

The idea of the method for finding an approximate solution to the problem (3), (4) for a slightly perturbed abrupt interface ($ka \ll 1$) is the following. Asymptotics (4) describe the wave field far from the interface, i.e., in the spatial domain $|z| \gg a$. On the other hand, the solution of (3) can be expanded into a series of k in the spatial domain $k|z|\sqrt{\varepsilon(z)-\cos^2\theta} \approx k|z| \ll 1$, or, in other words $|z| \ll \lambda$. If $ka \ll 1$, i.e., $a \ll \lambda$, the two domains intercept and the reflectivity R and transmittivity T in (4) can be found as a series of the wave number k:

$$R(k) = R_0 + kR_1 + k^2R_2 + ..., \tag{5}$$

$$T(k) = T_0 + kT_1 + k^2T_2 + ... \tag{6}$$

To illustrate the method, we will perform the derivation for the first correction R_1 only. Substituting (5) and (6) into the wave asymptotics (6), we get the following expressions which are valid, if $a \ll |z| \ll \lambda$:

$$E(z) = 1 + R_0 + k\left[R_1 + i(1-R_0)n_0z\right] + ..., \tag{7}$$

$$E'(z) = ikn_0(1-R_0) + k^2\left[-in_0R_1 - n_0^2(1+R_0)z\right] + ... \tag{8}$$

for $z < 0$, i.e., $-\lambda \ll z \ll -a$, and

$$E(z) = T_0 + k\left[T_1 + iT_0nz\right] + ..., \tag{9}$$

$$E'(z) = iknT_0 + k^2\left[inT_1 - n^2T_0z\right] + ... \tag{10}$$

for $z < 0$, i.e., $a \ll z \ll \lambda$.

On the other hand, for $|z| \ll \lambda$ the solution of (3) can be written as a series in powers of k:

$$E(z) = E_0(z) + kE_1(z) + k^2E_2(z) + ... \tag{11}$$

To determine $E_n(z)$ we substitute (11) into (3) and find

$$E_n'(z) = b_n - \int\limits_0^z \left[\varepsilon(z') - \cos^2\theta\right] E_{n-2}(z')dz', \tag{12}$$

$$E_n(z) = b_nz + c_n - \int\limits_0^z (z-z')\left[\varepsilon(z') - \cos^2\theta\right] E_{n-2}(z')dz', \tag{13}$$

where the constants b_n and c_n appear due to integration and

$$E_0(z) = b_0z + c_0, \quad E_1(z) = b_1z + c_1. \tag{14}$$

Formulas (11)–(14), as well as the set of expressions (7)–(10), represent the wave field for $a \ll |z| \ll \lambda$, therefore they should coincide.

Comparing (7) and (14) we obtain immediately that $b_0 = 0$. For a more detailed comparison, let us take formulas (12) and (13) at $|z| \gg \lambda$ and substitute them into (11) keeping only the first two nonvanishing terms in the fields $E(z)$ and $E'(z)$. Then we see that

$$E(z) = c_0 + k(b_1z + c_1) + ... \tag{15}$$

within all the domain $a \ll |z| \ll \lambda$, and

$$E'(z) = kb_1 - k^2\left[b_2 - c_0n_0^2z + c_0\int\limits_{-\infty}^{0}\left[\varepsilon(z) - 1\right]dz\right] + ..., \qquad -\lambda \ll z \ll -a, \tag{16}$$

$$E'(z) = kb_1 - k^2\left[b_2 - c_0n^2z - \int\limits_0^{\infty}\left[\varepsilon(z) - \varepsilon\right]dz\right] + ..., \qquad a \ll z \ll \lambda. \tag{17}$$

Now one can equalize the factors at k and k^2 in each pair of equations (7) and (15), (9) and (15), (8) and (16), and (10) and (17), as all of them represent the solution of the scattering problem (3), (4) in the same spatial domains. From the equations obtained one can find the involved quantities R_n, T_n, c_n, and b_n. In particular, we have

$$R_0 = R_{0s} = \frac{n_0 - n}{n_0 + n}, \quad R_1 = \frac{i(1+R_0)}{n_0+n} P, \quad P = \int\limits_{-\infty}^{\infty} \left[\varepsilon(z) - 1 - (\varepsilon - 1)\eta(z)\right] dz, \tag{18}$$

where $\eta(z)$ is a stepwise Heaviside function: $\eta(z) = 0$, $z < 0$ and $\eta(z) = 1$, $z > 0$.

Thus, we have found zero- and first-order terms in the expansion (5). Here R_0 is the Fresnel reflection coefficient from the ideally abrupt surface, whereas R_1 is the sought correction due to the smoothing of the spatial profile of permittivity $\varepsilon(z)$. Finally, according to (5) and (18) the reflectivity can be written as

$$R = R_0 \left(1 - \frac{2ik\sin\theta}{\varepsilon - 1} P\right) \tag{19}$$

with P given by (18).

3. P-polarization

The wave equation for p-polarization of the incident wave is usually written for the magnetic field B:

$$B'' - \frac{\varepsilon'(z)}{\varepsilon(z)} B' + \left[\varepsilon(z) - \cos^2\theta\right] B = 0. \tag{20}$$

The boundary conditions for B are the same as that for the electric field E [see (4)], and the reflection and transmission coefficients R_p and T_p are represented by the same series (5) and (6) in powers of k.[1] Therefore expansions (7)–(10) are valid for the magnetic field B too. Then, the general form (11) also holds true for the magnetic field B. However, it is easy to verify that the recurrent relations (12)–(14), due to the presence of the term $\frac{\varepsilon'}{\varepsilon} B$ in (20), are replaced by

$$B_n'(z) = \varepsilon(z) \left\{ b_n - \int\limits_0^z \left[1 - \frac{\cos^2\theta}{\varepsilon(z')}\right] B_{n-2}(z') dz' \right\}, \tag{21}$$

$$B_n(z) = F(z) b_n + c_n - \int\limits_0^z \left(F(z) - F(z')\right) \left[1 - \frac{\cos^2\theta}{\varepsilon(z')}\right] B_{n-2}(z') dz', \tag{22}$$

$$B_0(z) = b_0 F(z) + c_0, \quad B_1(z) = b_1 F(z) + c_1, \quad F(z) = \int\limits_0^z \varepsilon(z') dz'. \tag{23}$$

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[1] Further in Sec. 3 we omit indexes s and p for the sake of brevity.

Now, taking $|z| \gg a$ in (21)–(23) we obtain the following presentation of the magnetic field B within the domain $a \ll |z| \ll \lambda$:

$$B(z) = c_0 + k\left\{ b_1 z + c_1 - b_1 \int_{-\infty}^{0} [\varepsilon(z) - 1]\, dz \right\}, \tag{24}$$

$$B'(z) = kb_1 + k^2 \left\{ b_2 - c_0 z \sin^2\theta + c_0 \cos^2\theta \int_{-\infty}^{0} \left[1 - \frac{1}{\varepsilon(z)}\right] dz \right\} \tag{25}$$

for $-\lambda \ll z \ll -a$, and

$$B(z) = c_0 + k\left\{ b_1 \varepsilon z + c_1 + b_1 \int_{0}^{\infty} [\varepsilon(z) - \varepsilon]\, dz \right\}, \tag{26}$$

$$B'(z) = kb_1\varepsilon + k^2\varepsilon \left\{ b_2 - c_0 \cos^2\theta \int_{0}^{\infty} \left[\frac{1}{\varepsilon} - \frac{1}{\varepsilon(z)}\right] dz - c_0 \left(1 - \frac{\cos^2\theta}{\varepsilon(z)}\right) \right\} \tag{27}$$

for $a \ll z \ll \lambda$. Formulas (24)–(27), in a close analogy with Sec. 2, are to be compared with the expansions (7)–(10) (which, as was mentioned, are also true for the magnetic field). From this, we find again all the involved quantities R_n, T_n, c_n, and b_n including

$$R_0 = R_{0p} = \frac{n_0\varepsilon - n}{n_0\varepsilon + n}, \quad R_1 = -2in_0 R_0 \frac{(\varepsilon - \cos^2\theta)P + \varepsilon^2 \cos^2\theta P_1}{n_0^2\varepsilon^2 - n^2}, \tag{28}$$

where

$$P_1 = \int_{-\infty}^{\infty} \left[\frac{1}{\varepsilon(z)} - 1 - \left(\frac{1}{\varepsilon} - 1\right)\eta(z)\right] dz \tag{29}$$

and P is given by (18). The value R_0 in (28) is the Fresnel reflectivity at the ideally abrupt interface, and R_1 describes the correction connected with the smoothing of the spatial profile of permittivity $\varepsilon(z)$ [see expansion (5) which is valid for p-polarization too]. Thus, finally, we obtain the reflectivity of the p-polarized beam in the following form:

$$R = R_0 \left[1 - \frac{2ik\sin\theta}{\varepsilon - 1} P \frac{(\varepsilon - \cos^2\theta)P + \varepsilon^2\cos^2\theta P_1}{\varepsilon\sin^2\theta - \cos^2\theta}\right]. \tag{30}$$

4. Final Formulas

The use of (19) and (30) yields the following formulas for reflectivity $|R_s|^2$ and polarization factor $\rho = R_s/R_p$ of a sample covered by an overlayer:

$$|R_s(\theta)|^2 = |R_{0s}(\theta)|^2 (1 - A\sin\theta), \quad A = -4k\,\mathrm{Im}\frac{P}{\varepsilon - 1}, \tag{31}$$

$$\rho(\theta) = \rho_0(\theta)\left(1 - \frac{iA_1}{2}\frac{\sin\theta\cos^2\theta}{\varepsilon\sin^2\theta - \cos^2\theta}\right), \quad A_1 = \frac{4k\varepsilon}{\varepsilon - 1}(P + \varepsilon P_1), \tag{32}$$

where $R_{0s}(\theta)$ and $\rho_0(\theta)$ are the values given by the Fresnel formulas (1) and P and P_1 are determined by (18) and (29).

Thus, we express $|R_s|^2$ and $\rho(\theta)$ of a nonideal sample with graded interface in terms of the spatial integrals P and P_1 of the dielectric function $\varepsilon(z)$. If $\varepsilon(z)$ is a stepwise function $\varepsilon(z) = 1 + (\varepsilon - 1)\eta(z)$, then $A = A_1 = 0$ and no correction to the Fresnel formulas appear, as it should be. Formulas (31), (32) are valid within the accuracy of $a \sin\theta/\lambda$, where a is the scale of the overlayer thickness.

In the measurement of optical constants $\operatorname{Re}\varepsilon$ and $\operatorname{Im}\varepsilon$ from the reflectometry or ellipsometry data (when $\varepsilon(z)$ is usually unknown) the real value A and complex A_1 can be used as fitting parameters in formulas (31), (32) characterizing the contamination or transition layer at the surface of a sample.

5. Discussion

1. It can be shown directly from (3) and (20) that, if we replace $\varepsilon(z)$ by $\varepsilon(z-d)$, the reflectivities $R_s(\theta)$ and $R_p(\theta)$ acquire the factor $\exp(2ika\cos\theta)$ providing translation invariance, i.e., the observable characteristics $|R_s|^2$ and $\rho(\theta)$ of a sample do not depend on its location. One can verify that our result also satisfies this requirement. In other words, if we replace $\varepsilon(z)$ by $\varepsilon(z-d)$, the values $|R_s|^2$ and $\rho(\theta)$ given by (31), (32) stay unchanged.

2. Formula (31) satisfying general physical requirement of translation invariance is especially convenient for fitting the experimental data on bulk reflectivity. The contamination layer is effectively taken into account by only one real parameter A. Recall that asymptotically this is the exact result, if the thickness of the overlayer is small compared with the wavelength λ. Consequently, the smaller the value of A fitting the experimental reflectivity, the cleaner and more perfect the surface of the sample.

3. Fresnel formulas, as can be seen from (1) and (2), possess specific symmetry which is expressed by the relation

$$|R_{0s}(\theta)|^2 = \left|\frac{\rho_0(\theta) - \cos 2\theta}{1 - \rho_0(\theta)\cos 2\theta}\right|^2. \tag{33}$$

   Note that ε does not appear in (33). This relation does not hold for an arbitrary shape of the dielectric function and can be used as a check of abruptness of $\varepsilon(z)$. Using (31), (32) and taking into account (33), we have

$$\frac{1}{|R_s(\theta)|^2}\left|\frac{\rho(\theta) - \cos 2\theta}{1 - \rho(\theta)\cos 2\theta}\right|^2 = 1 + A\sin\theta + \frac{1}{4\sin\theta}\operatorname{Im}\left[\frac{A_1}{R_{0s}}\frac{(R_{0s} + \cos 2\theta)(1 + R_{0s}\cos 2\theta)}{\varepsilon\sin^2\theta - \cos^2\theta}\right]. \tag{34}$$

   Only values directly measured in reflectometry and ellipsometry are presented on the left-hand side of (34). Therefore, the latter also can be used for fitting the experimental data to find the bulk dielectric constant ε, as well as the overlayer parameters A and A_1.

6. Simulation

To check the accuracy of the formulas obtained above, several examples have been calculated.

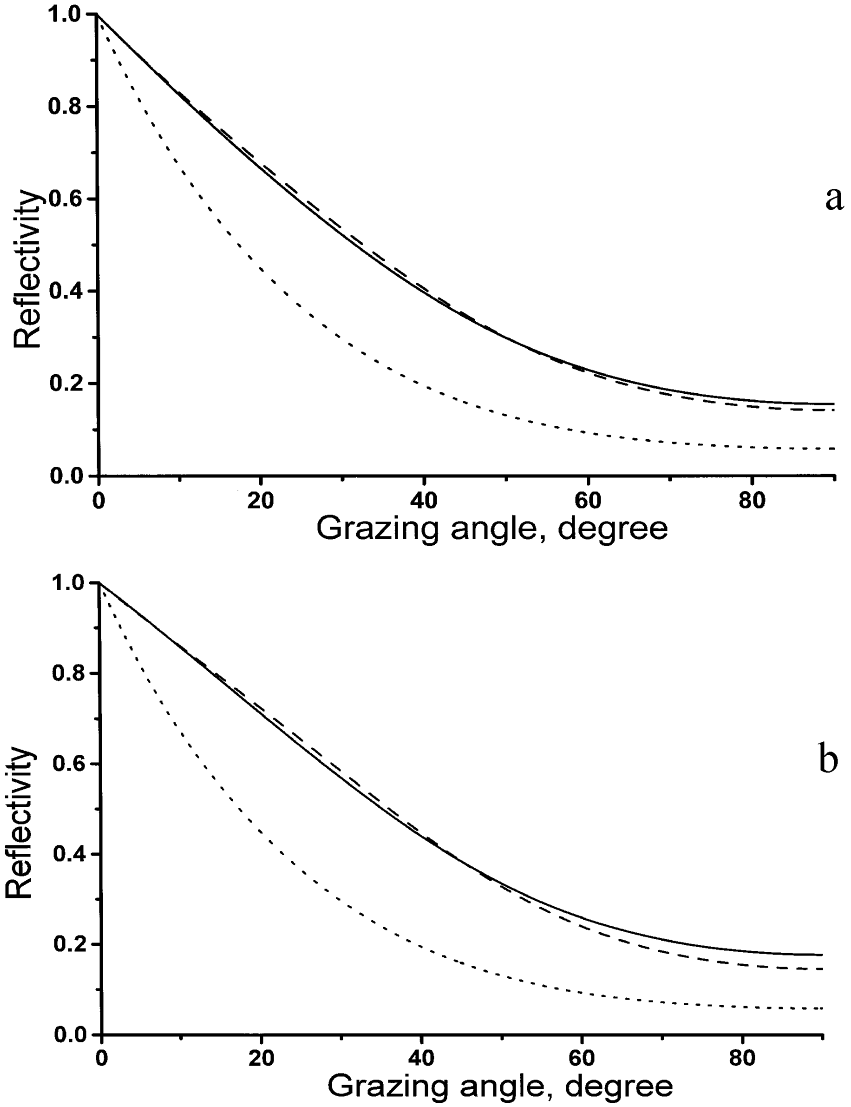

Fig. 1. Reflectivity of Ir covered with a contamination Ir film of thickness $d = 6.0$ nm (a) and 8.0 nm (b) at wavelength $\lambda = 46.9$ nm, s-polarization. The dashed and solid curves show the exact solution and approximate function (19), respectively, and the dotted curve corresponds to the case without a film.

The first deals with the wavelength 46.9 nm. The optical constants of various optical and microelectronic materials were determined in [5] on this wavelength using a compact capillary discharge laser. We calculated reflectivities using formulas (19) and (30) as well as the exact Fresnel expression assuming that all the interfaces are ideally abrupt. The results are shown in Figs. 1 and 2 for bulk Ir covered with a contamination film. Optical constants for all the materials were taken from [5]. One can see that our

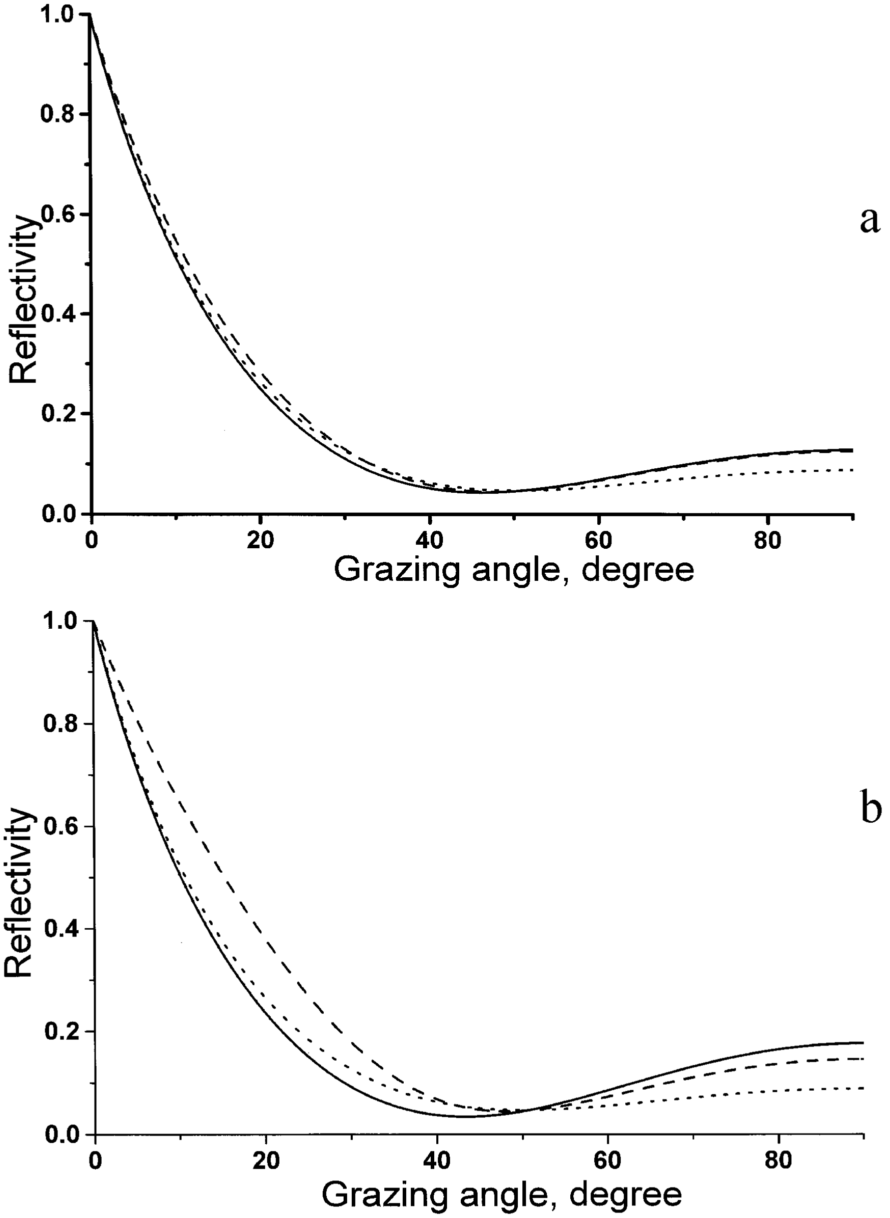

Fig. 2. Reflectivity of Ir covered with a contamination Ir film of thickness d = 3.6 nm (a) and 8.0 nm (b) at wavelength $\lambda = 46.9$ nm, p-plarization. Curves are the same as in Fig. 1.

approximation works for film thicknesses less than 8.0 nm.

The second example deals with the wavelength 13.0 nm that is important for VUVL lithography applications. The results are shown in Fig. 3 for bulk SiO_2 with C film. The calculation results for p-polarization are similar to those for s-polarization. Optical constants were taken from [9]. One can see that our approximation works for film thicknesses less than 4.0 nm.

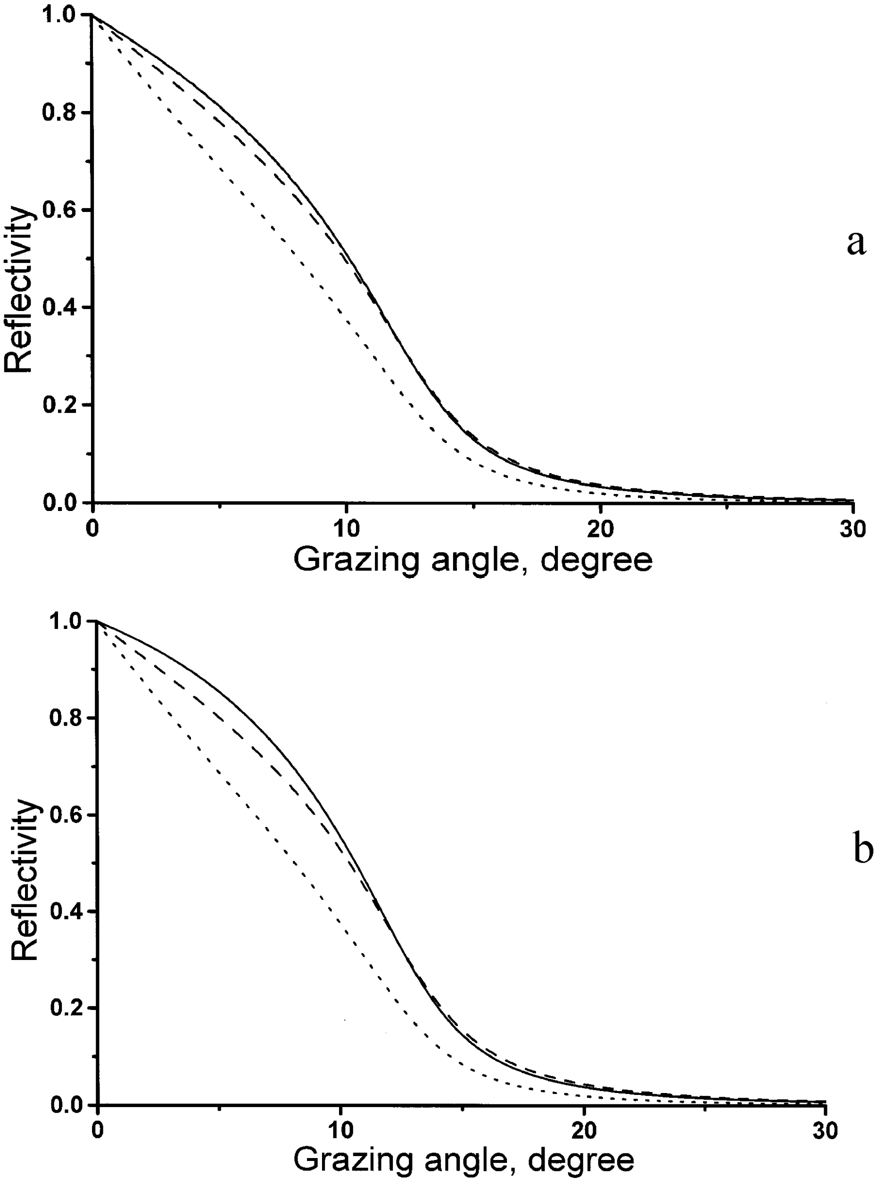

Fig. 3. Reflectivity of SiO_2 covered with a contamination C film of thickness $d = 3.0$ nm (a) and 4.0 nm (b) at wavelength $\lambda = 46.9$ nm, s-polarization. Curves are the same as in Fig. 1.

7. Summary

Formulas describing reflectometry and ellipsometry data in the presence of an overlayer on a bulk surface are suggested. They are based on general scattering theory and specific symmetry of the Fresnel reflection laws. The approach can be applied to improve the accuracy of determination of bulk optical constants, as well as to characterize the overlayer arising due to contamination or other interface imperfections. Model calculations show that in the XUV range it is capable of describing overlayers with thickness up to several nanometers.

Higher-order corrections to the Fresnel formulas proportional to $(a \sin\theta/\lambda)^2$ [6] can also be used. This can be important if the bulk and overlayer materials are both low absorbing. Our theoretical approach is an asymptotic solution of a differential wave equation. This method can be obviously extended to the case of a substrate coated by a film. The goal is to give an approximate description of the interfaces vacuum/film and film/bulk. This could be useful for measurement of optical constants of materials in thin films [8].

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