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To cite this article: Martin Čermák, Jiří Vohánka, Ivan Ohlídal & Daniel Franta (2018) Optical quantities of multi-layer systems with randomly rough boundaries calculated using the exact approach of the Rayleigh–Rice theory, Journal of Modern Optics, 65:14, 1720-1736

To link to this article: https://doi.org/10.1080/09500340.2018.1457187

Published online: 13 Jun 2018.

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Optical quantities of multi-layer systems with randomly rough boundaries calculated using the exact approach of the Rayleigh–Rice theory

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ABSTRACT
In this paper, the exact approach of the Rayleigh–Rice theory enabling us to calculate optical quantities of multi-layer systems with boundaries exhibiting slight random roughness is presented. This approach is exact in the sense that it takes into account the propagation of perturbed electromagnetic fields (waves) among randomly rough boundaries including all cross-correlation and auto-correlation effects. The restriction to the second order of perturbation, which is the lowest order that gives nonzero corrections to coherent waves (obeying the Snell's law), represents the only approximation used in our calculations. It is assumed that the layers and the substrates are formed by optically homogeneous and isotropic materials. The formulae obtained in the theoretical part are used to investigate the influence of layer thicknesses and roughness parameters on reflectances and associated ellipsometric parameters of the selected numerical examples of a three-layer system. The presented approach represents the generalization of the exact approach for single-layer systems and the improvement of the approximate approach for multi-layer systems published earlier. The exact approach of the RRT has a substantial importance for the optical characterization of multi-layer systems occurring in applied research and optics industry applications.

1. Introduction
Thin films occurring in practice exhibit many various defects influencing their properties. One of the most frequent defect is random boundary roughness. This roughness must be incorporated into formulae describing optical quantities of such the films if we want to perform their reliable and precise optical characterization. In the literature, several approaches were presented for including random boundary roughness. The usability of these approaches is especially dependent on magnitudes of heights and lateral dimensions of the roughness irregularities (1–3). If the characteristic heights and lateral dimensions of the irregularities are much smaller than the wavelength of incident light the effective medium approximation (EMA) is used to describe this roughness (4–17). If the lateral dimensions of boundary roughness are smaller, comparable or larger than the wavelength and heights are sufficiently smaller than this wavelength, perturbation theories such as the Rayleigh–Rice theory (RRT) can be used (9, 11, 18–27). If the randomly rough boundaries are locally smooth (roughness can locally be approximated by randomly tilted tangential planes in all points of the boundaries) one can use the scalar or vector diffraction theories (26, 28–46).

The optical characterization of multi-layer systems is on the rise (36, 37, 47, 48), therefore, the research of multi-layer systems with various defects is important. Practically all boundaries exhibit some degree of roughness. The current optical techniques using UV light are able to detect roughness with heights smaller than 1 nm. Therefore, for exact description of the system of thin layers the effects of rough boundaries must be included. The EMA, which can be used in similar type of boundary roughness as the RRT, gives quite good results for ellipsometry but fails when applied to spectrophotometry. In comparison to the EMA, the RRT gives more accurate results and, moreover, it is possible to determine the values of statistical parameters characterizing the rough boundaries. In the EMA, the effects of roughness are characterized by effective parameters which cannot be interpreted in this way. Using of the RRT appears to be the most accurate theory for describing the boundary roughness with small heights (compared to the wavelength of light) and small slopes. The disadvantage of RRT is that it is numerically demanding. The advances in the computer technology made it possible to use the exact approach of the RRT in practice.

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The RRT is the second-order perturbation theory used to calculate the optical quantities, i.e. reflectance, transmittance and ellipsometric parameters, corresponding to specularly reflected and transmitted light from randomly rough surfaces and layered systems. The RRT represents useful theoretical approach for the optical characterization of both the surfaces (18) and single layers (22). An application of the RRT to multi-layer systems exhibiting randomly rough boundaries has not been presented in a sufficient degree in the literature even when many rough multi-layer systems occur in practice. So far a simple approximate approach based on the RRT for including slight random boundary roughness into the formulae for the optical quantities of the multi-layer systems has only been published (25). This approximation is called the isolated roughness approximation (IRA). It is based on the assumption that the individual randomly rough boundaries are considered to be isolated at calculations of the optical quantities of the rough multi-layer system. This means that the usual matrix approach usable for the smooth multi-layer systems is used under assumption that the Fresnel coefficients of individual boundaries correspond to isolated randomly rough surfaces. It was shown that this approximation is satisfactory if the thicknesses of the individual layers forming layered systems are sufficiently large owing to lateral dimension of roughness (25). In this case it is possible to neglect the interaction of perturbed electromagnetic fields corresponding to the scattered waves from the rough boundaries inside the individual layers. For thinner layers this interaction can not be neglected and, therefore, this approximation can not be employed. Then one must formulate the exact approach taking into account the interaction of the scattered (perturbed) waves inside the system. In this paper, this problem will be solved exactly by means of the second-order RRT for the multi-layer system with the randomly rough boundaries. This means that the formulae for reflectances, transmittances and ellipsometric parameters of rough multi-layer systems taking into account the propagation of the perturbed waves will be derived for coherent waves. These formulae can then be utilized for the optical characterization of these systems. From the foregoing it is clear that our formulae derived here will represent the generalization of the approach derived for single-layer system (22) and they can replace approximate approach IRA presented in paper (25).

A brief numerical analysis of the formulae for reflectance and associated ellipsometric parameters of the multi-layer systems with slightly randomly rough boundaries will be presented in this paper in order to show the influence of the individual parameters of these systems on their optical quantities. Moreover, the comparison of the numerical results achieved by the new formulae to those obtained by the IRA is performed. By means of this comparison it is possible to estimate the limits of validity of the approximate approach concerning the multi-layer systems.

2. Theory

2.1. Specifications of the systems

In this paper, we will study system of \( q - 1 \) thin layers with \( q \) slightly rough boundaries. It is assumed that the mean planes of the boundaries are parallel. The Cartesian coordinate system is chosen such that the \( z \)-axis is oriented along the normals to the mean planes of the boundaries and the origin lies at the position of the mean plane of the upper boundary (see Figure 1). The boundaries are numbered \( i = 1, \ldots, q \) with \( i = 1 \) corresponding to the upper boundary and \( i = q \) corresponding to the lowest boundary. The media are numbered \( i = 0, \ldots, q \). The mean thickness of the layer formed by \( i \)-th media, i.e. the distance between the mean planes of the \( i \)-th and \( (i+1) \)-th boundary, is denoted by \( d_i \). The \( z \)-coordinate of the \( i \)-th boundary is given by

\[
z = f_i(x, y) - \sum_{j=1}^{i-1} d_j \equiv f_i(x, y) - h_i, \tag{1}
\]

where the function \( f_i(x, y) \) describes the deviations of the \( i \)-th boundary from its mean plane and \( h_i \) denotes the depth of this mean plane (note that the depth of the upper boundary is \( h_1 = 0 \)).

It is assumed that the deviations \( f_i(x, y) \) from the mean planes are small compared to wavelength of light and its first derivatives are small compared to unity. The vector perpendicular to the \( i \)-th boundary (see Figure 2) is given by...
where \( \mathbf{n} = (0, 0, 1) \) is the vector perpendicular to the mean planes of the boundaries. The subscripts \( ; x \) and \( ; y \) denote the partial derivatives with respect to coordinates \( x \) and \( y \).

At first it will be assumed that the functions \( f_i(x, y) \) are periodic in coordinates \( x \) and \( y \) with period \( L \), i.e. \( f_i(x, y) = f_i(x + L, y) = f_i(x, y + L) \). Therefore, this function can be expressed using the Fourier series as

\[
    f_i(x, y) = \sum_{m,n=-\infty}^{\infty} \hat{P}_{i,m,n} e^{i(Mx+Ny)},
\]

where \( \hat{P}_{i,m,n} \) are the Fourier coefficients; \( M = am, N = an \), where \( a = 2\pi/L \). The hat over \( \hat{P}_{i,m,n} \) is used to emphasize that these coefficients are complex numbers. Since \( f_i(x, y) \) is a real function, the condition \( \hat{P}_{i,-m,-n} = \hat{P}_{i,m,n}^* \) must hold.

The functions \( f_i(x, y) \) represent concrete realizations of the randomly rough boundaries. The mean values taken over different realizations will be denoted by the angled brackets. In the following calculations, the mean values of \( \hat{P}_{i,m,n} \) must be used \((22)\). The mean values of \( \hat{P}_{i,m,n} \) and \( \hat{P}_{i,m,n} \hat{P}_{j,k,l} \) for \( m \neq -k \), or \( n \neq -l \) vanish \((22)\)

\[
    \langle \hat{P}_{i,m,n} \hat{P}_{j,k,l} \rangle_{(m,n)\neq(-k,-l)} = 0.
\]

For \( m = -k, n = -l \) the mean value of \( \hat{P}_{i,m,n} \hat{P}_{j,k,l} \) is given by the power spectral density function (PSDF) \( \hat{W}_{i,j,m,n} \) as

\[
    \langle \hat{P}_{i,m,n} \hat{P}_{j,-m,-n} \rangle = a^2 \hat{W}_{i,j,m,n}.
\]

The PSDF describes distribution of spatial frequencies of the roughness.

As we will show, the corrections to the Fresnel coefficients from the first order of perturbation are proportional to \( \langle \hat{P}_{i,m,n} \rangle \) while the corrections from the second order are proportional to \( \langle \hat{P}_{i,m,n} \hat{P}_{j,k,l} \rangle \). Therefore, the corrections from the first order of perturbation to waves corresponding to specular reflection and refraction corresponding to the Snell’s law are zero. The second order of perturbation is the first order that gives non-vanishing corrections. If we studied the scattered light, then the first order of perturbation would be sufficient to get non-zero result.

### 2.2. Description of electromagnetic field

In the Rayleigh–Rice theory, the distribution of the electromagnetic field is given by the sum of unperturbed component, that corresponds to propagation of electromagnetic waves in the system with smooth boundaries, and corrections that take into account the influence of the rough boundaries. In this paper the corrections will be calculated up to the second perturbation order, which is the lowest order that gives non-zero corrections to the Fresnel reflection coefficients in the specular direction and transmission coefficients corresponding to Snell’s law.

In this work only non-magnetic \((\mu_r = 1)\), optically homogeneous and isotropic media will be considered. Moreover, it will be assumed that the ambient (0th medium) is non-absorbing.

#### 2.2.1. Unperturbed component of the electromagnetic field

As mentioned above the unperturbed component of the electromagnetic field corresponds to propagation of waves in the systems with smooth boundaries, i.e. assuming that the boundaries coincide with their mean planes. The electric field vector in the \( i \)th medium is a real part of \( \hat{E}_i^{(0)} \), where the index \( \beta \) distinguishes two types of polarizations: \( s \) (electric field vector is perpendicular to the plane of the incidence) or \( p \) (electric field vector is parallel with the plane of the incidence). The vectors \( \hat{E}_i^{(0)} \), at position \( \mathbf{r} = (x, y, z) \) and time \( t \) can be written as

\[
    \hat{E}_i^{(0)} = (0, 1, 0) e^{-i\omega t} \left[ \hat{r}_i^s e^{i\mathbf{k}_i^s \cdot \mathbf{r}} + \hat{r}_i^p e^{i\mathbf{k}_i^p \cdot \mathbf{r}} \right],
\]

\[
    \hat{E}_i^{p(0)} = e^{-i\omega t} \left[ \left( \cos \theta_i, 0, \sin \theta_i \right) \hat{r}_i^p e^{i\mathbf{k}_i^p \cdot \mathbf{r}} + \left( \cos \theta_i, 0, -\sin \theta_i \right) \hat{r}_i^p e^{i\mathbf{k}_i^p \cdot \mathbf{r}} \right],
\]

where \( \hat{r}_i^\beta \) and \( \hat{r}_i^p \) are the amplitudes of the waves propagating upwards (in the direction of \( z \)-axis) and downwards. The wave vectors are given by \( \mathbf{k}_i^\pm = \hat{k}_i \left( \sin \theta_i, 0, \mp \cos \theta_i \right) \), where \( \hat{k}_i = 2\pi \hat{n}_i/\lambda_0 \), the symbol \( \omega = 2\pi c/\lambda_0 \) denotes the angular frequency, \( \lambda_0 \) is wavelength of electromagnetic wave in vacuum and \( \hat{n}_i \) is the refractive index in the \( i \)th medium. The upper signs in \( \pm \) and \( \mp \) correspond
to waves propagating downwards and lower signs correspond to waves propagating upwards.

The symbols $\theta_i$, where $i \neq 0$, are the refraction angles and $\theta_0$ is the incidence angle. These angles must fulfill the Snell’s law

$$\hat{k}_i \sin \hat{\theta}_i = \hat{k}_{i-1} \sin \hat{\theta}_{i-1}. \quad (8)$$

If the electric field is known then the magnetic field can be calculated from the Maxwell equations as

$$\hat{B}_i^{(0)} = \hat{k}_i \frac{e^{-i\omega t}}{\omega} \left[ \left( \cos \hat{\theta}_i, 0, \sin \hat{\theta}_i \right) \hat{t}_i^* e^{i\hat{k}_i^* r} \right. \right.$$  
$$+ \left( - \cos \hat{\theta}_i, 0, \sin \hat{\theta}_i \right) \hat{r}_i^* e^{i\hat{k}_i^* r}, \quad (9)$$

$$\hat{B}_i^{(p)} = - (0, 1, 0) \hat{k}_i \frac{e^{-i\omega t}}{\omega} \left[ \hat{r}_i^* e^{i\hat{k}_i^* r} - \hat{\theta}_i^* e^{i\hat{k}_i^* r} \right]. \quad (10)$$

### 2.2.2. Perturbed component of the electromagnetic field

The electric and magnetic fields in the $i$th layer will be given as real parts of $\hat{E}_i^\beta$ and $\hat{B}_i^\beta$. These fields are given as a sum of unperturbed components, which were discussed in the previous section, and the corrections, which will be denoted by symbols $\hat{E}_i^\beta$ and $\hat{B}_i^\beta$. In the RRT the correction $\hat{E}_i^\beta$ to the electric field is written by means of the Fourier components $\hat{A}_{i,m,n}^\beta$, corresponding to the $x$ and $y$ directions. The indices $m$ and $n$ are the $x$ and $y$ components of the wave vector. The subscripts $+$ and $-$ are used to distinguish Fourier components corresponding to waves propagating downwards from those propagating upwards. Thus, the total electric field is given as

$$\hat{E}_i^\beta = \hat{E}_i^{\beta(0)} + \hat{E}_i^\beta = \hat{E}_i^{\beta(0)}$$
$$+ e^{-i\omega t} \sum_{m,n=-\infty}^{\infty} \hat{E}_{m,n} \left( \hat{A}_{i,m,n}^+ e^{-ib_{i,m,n}z} + \hat{A}_{i,m,n}^- e^{ib_{i,m,n}z} \right), \quad (11)$$

where $\hat{E}_{m,n} = e^{i(Mx+Ny)}$. The term $\hat{E}_{m,n}$ could be combined with exponential factors $e^{-i\omega t+ib_{i,m,n}z}$ into

$$\hat{E}_{m,n} e^{-i\omega t+ib_{i,m,n}z} = e^{-i(\omega t-\hat{k}_{i,m,n} r)}, \quad (12)$$

where $\hat{k}_{i,m,n} = (M, N, \mp b_{i,m,n})$ is the wave vector of the plane wave associated with the given Fourier coefficient $\hat{A}_{i,m,n}^\pm$. The square of the wave vector is equal to

$$\left( \hat{k}_{i,m,n}^\pm \right)^2 = M^2 + N^2 + b_{i,m,n}^2 = \frac{n_i^2}{\epsilon_i} \omega^2$$
$$= \left( \frac{2\pi n_i}{\lambda_0} \right)^2 = k_i^2. \quad (13)$$

From this equation one can express $\hat{b}_{i,m,n}$

$$\hat{b}_{i,m,n} = \sqrt{k_i^2 - M^2 - N^2}. \quad (14)$$

In order to ensure that the wave vector $\hat{K}_{i,m,n}^\pm$ corresponds to a wave propagating downwards, the square root in the above equation must be taken such that the result has non-negative imaginary part (and its real part is positive if imaginary part is zero). The vector $\hat{A}_{i,m,n}^\pm = (\hat{A}_{x,i,m,n}^\pm, \hat{A}_{y,i,m,n}^\pm, \hat{A}_{z,i,m,n}^\pm)$ describes the amplitude of a plane wave propagating in the $i$th medium in the direction determined by $\hat{K}_{i,m,n}^\pm$. The amplitudes of the waves incident on the system of layers with rough boundaries are completely determined by the unperturbed component. Thus, the amplitudes of the waves propagating downwards in the uppermost medium and the waves propagating upwards in the lowermost medium corresponding to the perturbed component must vanish

$$\hat{A}_{0,m,n}^+, \hat{A}_{q,m,n}^- = 0. \quad (15)$$

The magnetic field in the $i$th medium can be calculated from the electric field using the Maxwell equations

$$\hat{B}_i^\beta = \hat{B}_i^{\beta(0)} + \hat{B}_i^\beta = \hat{B}_i^{\beta(0)}$$
$$+ e^{-i\omega t} \sum_{m,n=-\infty}^{\infty} \hat{E}_{m,n} \left( \hat{A}_{i,m,n}^+ e^{-ib_{i,m,n}z} + \hat{A}_{i,m,n}^- e^{ib_{i,m,n}z} \right). \quad (16)$$

The condition of transversality of electromagnetic waves (Gauss law) is satisfied for the unperturbed waves and for the perturbed terms it gives the following conditions for the coefficients $\hat{A}_{i,m,n}$

$$\hat{K}_{i,m,n}^+ \cdot \hat{A}_{i,m,n}^+ = 0, \quad \hat{K}_{i,m,n}^- \cdot \hat{A}_{i,m,n}^- = 0. \quad (17)$$

Note that the condition of transversality of electromagnetic waves gives independent equation for each term in the Fourier expansion and direction of propagation, i.e., for given $M$ and $N$ the Equation (17) are $2q + 2$ independent equations.

### 2.3. Boundary conditions for unperturbed waves

The unperturbed component of the electromagnetic field is described by $\hat{E}_i^{\beta(0)}$ and $\hat{B}_i^{\beta(0)}$ (see Equations (6), (7), (9) and (10)). Since the unperturbed component corresponds to propagation of waves in the layered system with smooth boundaries the electric field and magnetic field in the nonmagnetic medium satisfy the following
normalised to unity then

\[ (\hat{E}_i^{\beta})^0 - \hat{E}_{i-1}^{\beta} ) \times \mathbf{n} = 0, \quad \left( \hat{B}_i^{\beta})^0 - \hat{B}_{i-1}^{\beta} \right) \times \mathbf{n} = 0, \]

where \( z = -h_i \). Boundary conditions can be solved separately for each polarization. Substituting Equations (6), (7), (9), (10) into these boundary conditions we get

\[ \hat{\eta}_i^e (\hat{\eta}_i^e + \hat{\eta}_i^m) = \hat{\eta}_{i-1}^e - \hat{\eta}_{i-1}^m, \]

\[ \hat{\eta}_i^m (\hat{\eta}_i^e - \hat{\eta}_i^m) = \hat{\eta}_{i-1}^m - \hat{\eta}_{i-1}^e, \]

\[ \hat{\eta}_i^e = \hat{\eta}_{i-1}^e - \hat{\eta}_{i-1}^m, \]

\[ \hat{\eta}_i^m = \hat{\eta}_{i-1}^m - \hat{\eta}_{i-1}^e. \]

In this section, the boundary conditions for the electromagnetic fields at rough boundaries will be derived. If the electric and magnetic fields are separated into unperturbed component and the correction as indicated in (11) and (16) then the boundary conditions at \( t = 0 \) boundary can be written as

\[ \left( \hat{E}_i^{\beta} - \hat{E}_{i-1}^{\beta} \right) \times \mathbf{n}_i = \mathbf{n}_i \times \left( \hat{E}_i^{\beta})^0 - \hat{E}_{i-1}^{\beta} \right), \]

\[ \left( \hat{B}_i^{\beta} - \hat{B}_{i-1}^{\beta} \right) \times \mathbf{n}_i = \mathbf{n}_i \times \left( \hat{B}_i^{\beta})^0 - \hat{B}_{i-1}^{\beta} \right). \]

The vector of amplitudes \( \hat{\sigma}^\beta \) can be easily expressed from Equation (24) as

\[ (\hat{\sigma})^\beta = (\hat{\mu}^\beta)^{-1} \mathbf{v}. \]
The system of equations following from (31) and (32) is not linearly independent, it is enough to consider only the x and y components of these boundary conditions, i.e., the x and y components of the vectors \( \vec{\tau}_{i,L}^β, \vec{\tau}_{i,R}^β, \vec{\phi}_{i,L}^β \) and \( \vec{\phi}_{i,R}^β \).

The x and y components of the vector \( \vec{\tau}_{i,L}^β \) are calculated using (11) and (33)

\[
\vec{\tau}_{i,x,L}^β = \sum_{m,n=-\infty}^{\infty} \hat{E}_{m,n} \left\{ \left[ (1 - i\beta_{i,m,n}) \right] \hat{A}_{x_{i,m,n}}^{β} + \hat{A}_{z_{i,m,n}}^{β} e^{i\phi_{i,m,n}} ight. \\
+ \left[ (1 + i\beta_{i,m,n}) \right] \hat{A}_{y_{i,m,n}}^{β} e^{-i\phi_{i,m,n}} \\
- \left[ (1 - i\beta_{i,m,n}) \right] \hat{A}_{y_{i,m,n}}^{β} e^{i\phi_{i,m,n}} \\
+ \left[ (1 + i\beta_{i,m,n}) \right] \hat{A}_{x_{i,m,n}}^{β} e^{-i\phi_{i,m,n}} \right\}.
\]

(34)

\[
\vec{\tau}_{i,y,L}^β = \sum_{m,n=-\infty}^{\infty} \hat{E}_{m,n} \left\{ \left[ (1 - i\beta_{i,m,n}) \right] \hat{A}_{y_{i,m,n}}^{β} + \hat{A}_{z_{i,m,n}}^{β} e^{i\phi_{i,m,n}} \\
+ \left[ (1 + i\beta_{i,m,n}) \right] \hat{A}_{x_{i,m,n}}^{β} e^{-i\phi_{i,m,n}} \\
- \left[ (1 - i\beta_{i,m,n}) \right] \hat{A}_{x_{i,m,n}}^{β} e^{i\phi_{i,m,n}} \\
+ \left[ (1 + i\beta_{i,m,n}) \right] \hat{A}_{y_{i,m,n}}^{β} e^{-i\phi_{i,m,n}} \right\}.
\]

(35)

While the electric and magnetic fields \( \hat{E}_i^β \) and \( \hat{B}_i^β \) of the left hand sides of (31) and (32) are at least the first order of perturbation, the unperturbed fields \( \hat{E}_i^{β(0)} \) and \( \hat{B}_i^{β(0)} \) on the right hand sides represent the zeroth order of the perturbation theory. For this reason it is not enough to expand the exponential factors using the first-order Taylor series and the expansion to the second order must be used, i.e.

\[
e^{ik_i^β r_i} \equiv e^{ik_i^β r_i} \left[ 1 + i k_f^β \cos \hat{\theta}_i - \frac{1}{2} (k_f^β \cos \hat{\theta}_i)^2 \right],
\]

\[
e^{ik_{i-1}^β r_i} \equiv e^{ik_{i-1}^β r_i} \left[ 1 + i k_{f_{i-1}}^β \cos \hat{\theta}_{i-1} - \frac{1}{2} (k_{f_{i-1}}^β \cos \hat{\theta}_{i-1})^2 \right],
\]

(36)

where \( r_i^β = (x_i, y_i, f_i - h_i) \) and \( r_i^0 = (x_i, y_i, -h_i) \). With the help of Equations (8), (36), (19) and (20) the x and y components of the vector \( \vec{\tau}_{i,R}^β \) for s polarization are expressed as

\[
\vec{\tau}_{i,x,R}^s = \frac{f_i^2}{2} \left( \hat{k}_{i-1}^2 - \hat{k}_{i}^2 \right) \left( \hat{t}_{i-1}^s e^{i\hat{k}_{i-1}^β r_i^s} + \hat{t}_{i-1}^s e^{-i\hat{k}_{i-1}^β r_i^s} \right),
\]

\[
\vec{\tau}_{i,y,R}^s = 0.
\]

(37)

Note that the terms proportional to the zeroth and the first terms in the Taylor series (36) do not contribute to this results due to the identities (19) and (20). For further calculations it is convenient to write one of the functions \( f_i \) in (37) using the Fourier components \( \hat{P}_{i,m,n} \) (see (3)) as

\[
\vec{\tau}_{i,x,R}^s = \sum_{m,n=-\infty}^{\infty} \hat{P}_{i,m,n} \hat{E}_{m,n} \frac{f_i}{2} \left( \hat{k}_{i-1}^2 - \hat{k}_{i}^2 \right) \times \left( \hat{t}_{i-1}^s e^{i\hat{k}_{i-1}^β r_i^s} + \hat{t}_{i-1}^s e^{-i\hat{k}_{i-1}^β r_i^s} \right).
\]

(38)

The expressions in the exponentials can be written as \( ik_i^β r_i^β = iaν x ± \hat{\phi}_i \), where \( aν = k_i \sin \hat{\theta}_i = k_0 \sin \hat{\theta}_o \). It is possible to perform a substitution \( m = m' + ν \) and the term \( \vec{\tau}_{i,x,R}^s \) can be written as

\[
\vec{\tau}_{i,x,R}^s = \sum_{m,n=-\infty}^{\infty} \hat{P}_{i,m-n,v} \hat{E}_{m,n} \frac{f_i}{2} \left( \hat{k}_{i-1}^2 - \hat{k}_{i}^2 \right) \times \left( \hat{t}_{i-1}^s e^{i\hat{\phi}_i} + \hat{t}_{i-1}^s e^{-i\hat{\phi}_i} \right).
\]

(39)

The similar steps to those performed for the s polarization can be used to find the x and y components of the vector \( \vec{\tau}_{i,R}^p \) for the p polarization. The result is as follows

\[
\vec{\tau}_{i,x,R}^p = -iν \sum_{m,n=-\infty}^{\infty} \hat{P}_{i,m-v,n} \hat{E}_{m,n} \left( \hat{t}_{i-1}^p e^{i\hat{\phi}_i} - \hat{t}_{i-1}^p e^{i\hat{\phi}_i} \right),
\]

\[
\times \left( \sin \hat{\theta}_{i-1} - \sin \hat{\theta}_{i-1} \right),
\]

(40)

\[
\vec{\tau}_{i,y,R}^p = \sum_{m,n=-\infty}^{\infty} \hat{P}_{i,m-v,n} \hat{E}_{m,n} \left( \frac{f_i}{2} \cos \hat{\theta}_{i-1} \left( \hat{k}_{i-1}^2 - \hat{k}_{i}^2 \right) \times \left( \hat{t}_{i-1}^p e^{i\hat{\phi}_i} + \hat{t}_{i-1}^p e^{-i\hat{\phi}_i} \right) + i \left( \hat{t}_{i-1}^p e^{i\hat{\phi}_i} - \hat{t}_{i-1}^p e^{-i\hat{\phi}_i} \right) \right) \times \left( M - aν \right) \sin \hat{\theta}_{i-1} \left( \frac{\sin \hat{\phi}_{i-1}^r - \sin \hat{\phi}_{i-1}^r}{k_i} \right) - i\hat{k}_{i-1} \left( \cos \hat{\theta}_{i-1}^r - \cos \hat{\theta}_{i-1}^r \right) \right).\]

(41)

The expressions on the left-hand side of the boundary conditions for the magnetic field (18) can be treated in the same way as those for the electric field, i.e. the magnetic fields are expressed as (16) and the exponential factors are expanded into the second-order the Taylor series (33). If
all the terms of the perturbation order higher than two are neglected the following results are obtained

$$\hat{\sigma}_{i,x,L} = \sum_{m,n=-\infty}^{\infty} E_{m,n} \left[ -\hat{b}_{1,m,n} \left( 1 - i b_{1,m,n} \right) \hat{A}_{x,i,m,n}^{+} \right. $$

$$+ M \left( 1 - i b_{1,m,n} \right) \hat{A}_{x,i,m,n}^{+} $$

$$+ f_{i,y} \hat{M}_{y,i,m,n} \hat{A}_{x,i,m,n}^{+} $$

$$- f_{i,y} \hat{N}_{a,i,m,n} \hat{A}_{x,i,m,n}^{+} $$

$$+ \hat{b}_{1,m,n} \left( 1 + i b_{1,m,n} \right) \hat{A}_{y,i,m,n}^{+} $$

$$- M \left( 1 + i b_{1,m,n} \right) \hat{A}_{y,i,m,n}^{+} $$

$$+ f_{i,y} \hat{M}_{y,i,m,n} \hat{A}_{y,i,m,n}^{+} $$

$$- f_{i,y} \hat{N}_{a,i,m,n} \hat{A}_{y,i,m,n}^{+} $$

$$- \left[ -\hat{b}_{1,m,n} \left( 1 - i b_{1,m,n} \right) \hat{A}_{x,i+1,m,n}^{+} \right. $$

$$- M \left( 1 - i b_{1,m,n} \right) \hat{A}_{x,i+1,m,n}^{+} $$

$$+ f_{i,y} \hat{M}_{y,i+1,m,n} \hat{A}_{x,i+1,m,n}^{+} $$

$$- f_{i,y} \hat{N}_{a,i+1,m,n} \hat{A}_{x,i+1,m,n}^{+} $$

$$- \left[ -\hat{b}_{1,m,n} \left( 1 + i b_{1,m,n} \right) \hat{A}_{y,i+1,m,n}^{+} \right. $$

$$- M \left( 1 + i b_{1,m,n} \right) \hat{A}_{y,i+1,m,n}^{+} $$

$$+ f_{i,y} \hat{M}_{y,i+1,m,n} \hat{A}_{y,i+1,m,n}^{+} $$

$$- f_{i,y} \hat{N}_{a,i+1,m,n} \hat{A}_{y,i+1,m,n}^{+} \left] \right. $$

$$\left. \sum_{m,n=-\infty}^{\infty} \hat{P}_{l,m,n} \hat{E}_{m,n} \right] $$

(42)

$$\hat{\sigma}_{i,y,R} = \sum_{m,n=-\infty}^{\infty} \hat{P}_{l,m,n} \hat{E}_{m,n} $$

$$\times \left[ \hat{k}_{n}^{2} - \hat{k}_{i,n}^{2} \right] \left[ -i \cos \hat{b}_{i,n} \left( \hat{P}_{l,n} e^{i \hat{\psi}} + \hat{P}_{l,n} e^{-i \hat{\psi}} \right) \right. $$

$$- \frac{f_{i,n}}{2} \hat{k}_{i,n} \left( \hat{P}_{l,n} e^{i \hat{\psi}} - \hat{P}_{l,n} e^{-i \hat{\psi}} \right) \left] \right. $$

(44)

$$\hat{\sigma}_{i,x,R} = \sum_{m,n=-\infty}^{\infty} \hat{P}_{l,m,n} \hat{E}_{m,n} $$

$$\times \left[ \hat{k}_{n}^{2} - \hat{k}_{i,n}^{2} \right] \left[ -i \left( \hat{P}_{l,n} e^{i \hat{\psi}} + \hat{P}_{l,n} e^{-i \hat{\psi}} \right) \right. $$

$$- \frac{f_{i,n}}{2} \hat{k}_{i,n} \cos \hat{b}_{i,n} \left( \hat{P}_{l,n} e^{i \hat{\psi}} - \hat{P}_{l,n} e^{-i \hat{\psi}} \right) \right] \left. \right] $$

(45)

and the other components of the vectors $\hat{\sigma}_{i,R}$ are equal to zero, i.e. $\hat{\sigma}_{i,y,R} = 0$, $\hat{\sigma}_{i,x,R} = 0$.

### 2.5. First order of perturbation calculations

The boundary conditions derived in the previous section are written up to the second order of the perturbation. Let us remind, that in our perturbation calculations the functions $f_{i,x}$ and their derivatives $f_{i,x}$, $f_{i,y}$ are considered to be the first-order terms. So far the electric and magnetic fields were separated into the unperturbed components $E_{i,0}$, $B_{i,0}$, which correspond to the zeroth order of the perturbation theory, and the corrections $E_{i,1}$, $B_{i,1}$, which include the first and higher orders. In order to solve the boundary conditions perturbatively it is necessary to write the amplitudes $A_{i,m,n}$, which are used to express the corrections $E_{i,1}$ (11) and $B_{i,1}$ (16), as a sum of terms corresponding to different orders of the perturbation theory

$$\hat{A}_{i,m,n} = \hat{A}_{i,m,n}^{\pm}(1) + \hat{A}_{i,m,n}^{\pm}(2) + \cdots $$

(46)

where $\hat{A}_{i,m,n}^{\pm}(1)$ and $\hat{A}_{i,m,n}^{\pm}(2)$ represent the first- and second-order terms, respectively.

After inserting the perturbation expansion (46) into (34), (35), (42) and (43) one can split the results according to perturbation order. In the first-order terms the dependency on the coordinates $x$ and $y$ is only through the terms $\hat{E}_{m,n}$. Therefore, the first perturbation order of the boundary conditions can be written by means of the Fourier expansion as

$$\hat{\tau}_{l,x,L}^{(1)} = \sum_{m,n=-\infty}^{\infty} \hat{E}_{m,n} \hat{\tau}_{l,m,n}^{(1)}, \quad \hat{\tau}_{l,y,L}^{(1)} = \sum_{m,n=-\infty}^{\infty} \hat{E}_{m,n} \hat{\tau}_{l,m,n}^{(1)},$$

(47)
where the superscript (1) is used to denote the first-order terms and \( \hat{r}_{i,m,n,L}^{(1)}, \hat{r}_{i,m,n,R}^{(1)}, \hat{\sigma}_{i,m,n,L}^{(1)}, \hat{\sigma}_{i,m,n,R}^{(1)} \) are the Fourier coefficients. From the equalities between the left and right sides of the boundary conditions (31) and (32) it follows that the corresponding Fourier coefficients must be equal

\[
\hat{r}_{i,m,n,L}^{(1)} = r_{i,m,n,L}^{(1)}, \quad \hat{r}_{i,m,n,R}^{(1)} = r_{i,m,n,R}^{(1)}, \quad \hat{\sigma}_{i,m,n,L}^{(1)} = \sigma_{i,m,n,L}^{(1)}, \quad \hat{\sigma}_{i,m,n,R}^{(1)} = \sigma_{i,m,n,R}^{(1)}. \tag{48}
\]

The Fourier coefficients \( \hat{r}_{i,m,n,L}^{(1)}, \hat{r}_{i,m,n,R}^{(1)}, \hat{\sigma}_{i,m,n,L}^{(1)}, \hat{\sigma}_{i,m,n,R}^{(1)} \) are obtained from (34), (35), (42) and (43)

\[
\begin{align*}
\hat{r}_{i,x,m,n,L}^{(1)} &= \hat{A}_{x,i,m,n} e^{i \beta_{i,m,n} h_i} + \hat{\beta}_{x,i,m,n} e^{-i \beta_{i,m,n} h_i} \\
\hat{r}_{i,x,m,n,R}^{(1)} &= \hat{A}_{y,i,m,n} e^{-i \beta_{i,m,n} h_i} + \hat{\beta}_{y,i,m,n} e^{i \beta_{i,m,n} h_i} \\
\hat{\sigma}_{i,y,m,n,L}^{(1)} &= -\hat{A}_{x,i,m,n} e^{i \beta_{i,m,n} h_i} + \hat{\beta}_{x,i,m,n} e^{-i \beta_{i,m,n} h_i} \\
\hat{\sigma}_{i,y,m,n,R}^{(1)} &= -\hat{A}_{y,i,m,n} e^{-i \beta_{i,m,n} h_i} + \hat{\beta}_{y,i,m,n} e^{i \beta_{i,m,n} h_i}.
\end{align*}
\tag{49}
\]

Apart from the boundary conditions, the amplitudes \( \hat{A}_{i,m,n} \) must fulfill the condition of transversality of electromagnetic waves (17)

\[
\begin{align*}
0 &= \hat{A}_{x,i,m,n} M + \hat{A}_{y,i,m,n} N - \hat{A}_{z,i,m,n} \hat{b}_{i-1,m,n} \\
0 &= \hat{A}_{x,i-1,m,n} M + \hat{A}_{y,i-1,m,n} N + \hat{A}_{z,i-1,m,n} \hat{b}_{i-1,m,n}
\end{align*}
\tag{51}
\]

and it is also known that the amplitudes of the incident waves are completely determined in the zeroth order of perturbation, thus, the first-order contributions to these amplitudes must be zero

\[
\begin{align*}
\hat{A}_{x,i,m,n,0} &= 0, \quad \hat{A}_{y,i,m,n,0} = 0, \quad \hat{A}_{z,i,m,n,0} = 0, \\
\hat{A}_{x,i,m,n,q} &= 0, \quad \hat{A}_{y,i,m,n,q} = 0, \quad \hat{A}_{z,i,m,n,q} = 0.
\end{align*}
\tag{52}
\]

The Equations (48) and (51) with \( i = 1, \ldots, q \) together with (52) form a system of 6\( q \) + 6 independent linear equations that can be used to find unknown amplitudes \( \hat{A}_{i,m,n} \). This family of linear equations can be rewritten in the matrix form as

\[
\begin{pmatrix}
\hat{M}_{m,n} \hat{A}_{m,n} \\
\hat{R}_{m,n}
\end{pmatrix} = i \hat{P}_{m,v,n} \hat{R}_{m,n},
\tag{53}
\]

where the vectors of amplitudes \( \hat{A}_{m,n} \), vectors \( \hat{R}_{m,n} \) on the right hand side, diagonal matrix \( \hat{P}_{m,v,n} \) and the matrix \( \hat{M}_{m,n} \) are given by

\[
\begin{align*}
\hat{A}_{m,n} &= \begin{pmatrix}
\hat{A}_{m,n,0} & \hat{A}_{m,n,1} & \cdots & \hat{A}_{m,n,q}
\end{pmatrix}^T, \\
\hat{R}_{m,n} &= \begin{pmatrix}
\hat{R}_{m,n,0} & \hat{R}_{m,n,1} & \cdots & \hat{R}_{m,n,q}
\end{pmatrix}^T, \\
\hat{P}_{m,v,n} &= \begin{pmatrix}
\hat{P}_{m,v,n,0} & 0 & 0 & 0 & \cdots
\end{pmatrix}
\end{align*}
\tag{54}
\tag{55}
\tag{56}
\]

The Equations (48) and (51) with \( i = 1, \ldots, q \) together with (52) form a system of 6\( q \) + 6 independent linear equations that can be used to find unknown amplitudes \( \hat{A}_{i,m,n} \). This family of linear equations can be rewritten in the matrix form as

\[
\begin{pmatrix}
\hat{A}_{m,n} \\
\hat{R}_{m,n}
\end{pmatrix} = i \hat{P}_{m,v,n} \hat{R}_{m,n},
\tag{53}
\]

where the vectors of amplitudes \( \hat{A}_{m,n} \), vectors \( \hat{R}_{m,n} \) on the right hand side, diagonal matrix \( \hat{P}_{m,v,n} \) and the matrix \( \hat{M}_{m,n} \) are given by

\[
\hat{A}_{m,n} = \begin{pmatrix}
\hat{A}_{m,n,0} & \hat{A}_{m,n,1} & \cdots & \hat{A}_{m,n,q}
\end{pmatrix}^T, \\
\hat{R}_{m,n} = \begin{pmatrix}
\hat{R}_{m,n,0} & \hat{R}_{m,n,1} & \cdots & \hat{R}_{m,n,q}
\end{pmatrix}^T, \\
\hat{P}_{m,v,n} = \begin{pmatrix}
\hat{P}_{m,v,n,0} & 0 & 0 & 0 & \cdots
\end{pmatrix}
\tag{54}
\tag{55}
\tag{56}
\]
In this section, the second-order corrections to the electric field amplitudes will be calculated. In order to simplify our calculations we will split the second-order terms on the left hand sides of the boundary conditions (31) and (32) into two parts. The first part \( \hat{\beta}_{i,L}^{(2a)} \) and \( \hat{\beta}_{i,L}^{(2a)} \) will consist of terms proportional to the second-order corrections to the amplitudes \( \hat{A}_{i,m,n}^{(2)} \) while the second part \( \hat{\beta}_{i,L}^{(2b)} \) and \( \hat{\beta}_{i,L}^{(2b)} \) will be proportional to the first-order corrections to the amplitudes \( \hat{A}_{i,m,n}^{(1)} \). The second-order terms on the right-hand sides of (31) and (32) will be denoted by the symbols \( \hat{\beta}_{i,L}^{(2)} \) and \( \hat{\beta}_{i,L}^{(2)} \). Thus, the boundary conditions for the second order of perturbation can be written as

\[
\hat{\beta}_{i,L}^{(2a)} = \hat{\beta}_{i,L}^{(2a)} - \hat{\beta}_{i,L}^{(2b)}, \quad \hat{\beta}_{i,L}^{(2b)} = \hat{\beta}_{i,L}^{(2b)} - \hat{\beta}_{i,L}^{(2b)}.
\]  

(62)

The amplitudes \( \hat{A}_{i,m,n}^{(2)} \) appear only on the left hand side of these equations. The system of linear equations that can be solved to obtain these amplitudes can be expressed in the matrix form similarly as was done for the first-order amplitudes in (53). The result for the left hand side can be written as

\[
\sum_{k,l=-\infty}^{\infty} \hat{E}_{k,l} \hat{M}_{k,l} \hat{A}_{k,l}^{(2)} = 0.
\]

(63)

where the matrix \( \hat{M}_{k,l} \) (59) is the same matrix as the one used in the first order of the perturbation theory. The vector of amplitudes \( \hat{A}_{k,l}^{(2)} \) is defined as

\[
\hat{A}_{k,l}^{(2)} = \left( \hat{A}_{k,l,0}^{(2)}, \hat{A}_{k,l,0}^{(2)}, \hat{A}_{k,l,1}^{(2)}, \hat{A}_{k,l,1}^{(2)}, \ldots, \hat{A}_{k,l,q}^{(2)} \right)^T.
\]

(64)

Note that the matrix \( \hat{M}_{k,l} \) includes the condition of transversality of electromagnetic waves and the conditions that the second-order contributions to the amplitudes of the waves incident on the system in question are zero, i.e. it includes the left hand sides of the following equations

\[
\hat{\beta}_{i,m,n}^{(2)} = \hat{\beta}_{i,m,n}^{(2)} - \hat{\beta}_{i,m,n}^{(2)} = 0,
\]

\[
\hat{\beta}_{i,m,n}^{(2)} = \hat{\beta}_{i,m,n}^{(2)} - \hat{\beta}_{i,m,n}^{(2)} = 0.
\]

(65)

The second-order terms \( \hat{\beta}_{i,R}^{(2)} \) and \( \hat{\beta}_{i,R}^{(2)} \), which are proportional to the amplitudes of the unperturbed wave,
can be obtained from (39), (41), (44) and (45). They are as follows

\[ \tilde{r}^{s(2)}_{i,x,R} = \sum_{n,m=-\infty}^{\infty} \hat{E}_{m,n} \hat{P}_{i,m-n} \frac{f_i}{2} \left( \tilde{k}_i^2 - \tilde{k}_{i-1}^2 \right) \times \left( \tilde{t}^s_{i-1} e^{\tilde{\varphi}_i} + \tilde{r}^s_{i-1} e^{-\tilde{\varphi}_i} \right), \]

\[ \tilde{r}^{s(2)}_{i,y,R} = 0, \]

\[ \tilde{r}^{p(2)}_{i,x,R} = 0, \]

\[ \tilde{r}^{p(2)}_{i,y,R} = - \sum_{n,m=-\infty}^{\infty} \hat{E}_{m,n} \hat{P}_{i,m-n} \frac{f_i}{2} \cos \tilde{\theta}_{i-1} \left( \tilde{k}_i^2 - \tilde{k}_{i-1}^2 \right) \times \left( \tilde{t}^p_{i-1} e^{\tilde{\varphi}_i} + \tilde{r}^p_{i-1} e^{-\tilde{\varphi}_i} \right), \]

\[ \tilde{r}^{s(2)}_{i,x,R} = 0, \]

\[ \tilde{r}^{s(2)}_{i,y,R} = - \sum_{n,m=-\infty}^{\infty} \hat{E}_{m,n} \hat{P}_{i,m-n} \left( \tilde{k}_i^2 - \tilde{k}_{i-1}^2 \right) \times \frac{f_i}{2} \tilde{\theta}_{i-1} \sin \tilde{\theta}_{i-1} \left( \tilde{t}^s_{i-1} e^{\tilde{\varphi}_i} - \tilde{r}^s_{i-1} e^{-\tilde{\varphi}_i} \right), \]

\[ \tilde{r}^{p(2)}_{i,x,R} = - \sum_{n,m=-\infty}^{\infty} \hat{E}_{m,n} \hat{P}_{i,m-n} \left( \tilde{k}_i^2 - \tilde{k}_{i-1}^2 \right) \times \frac{f_i}{2} \tilde{\theta}_{i-1} \sin \tilde{\theta}_{i-1} \left( \tilde{t}^p_{i-1} e^{\tilde{\varphi}_i} - \tilde{r}^p_{i-1} e^{-\tilde{\varphi}_i} \right), \]

\[ \tilde{r}^{s(2)}_{i,y,R} = 0. \]

All of the above expressions consist of a sum over \( m \) and \( n \) of terms proportional to the product of \( \hat{E}_{m,n}, \hat{P}_{i,m-n} \) and \( f_i \). For this reason it is convenient to separate this common part and organize the remaining part into the six-dimensional vectors \( \tilde{r}_{i,m,n}^{(2)} \) defined such that

\[ \sum_{n,m=-\infty}^{\infty} \hat{E}_{m,n} f_i \hat{P}_{i,m-n} \tilde{r}_{i,m,n}^{(2)} = \left( 0, \tilde{r}^{H(2)}_{i,x,R} \right), \]

\[ \sum_{n,m=-\infty}^{\infty} \hat{E}_{m,n} f_i \hat{P}_{i,m-n} \tilde{r}_{i,m,n}^{(2)} = \left( 0, \tilde{r}^{H(2)}_{i,y,R} \right). \]

The functions \( f_i \) on the left hand sides of the above equations can be expanded into the Fourier series (3) and the result can be written as follows

\[ \sum_{n,m=-\infty}^{\infty} \hat{E}_{m,n} f_i \hat{P}_{i,m-n} \tilde{r}_{i,m,n}^{(2)} = \sum_{n,m'=-\infty}^{\infty} \hat{E}_{m+n,m',n'} \hat{P}_{i,m-n} \tilde{r}_{i,m,n}^{(2)} = \sum_{k,l=-\infty}^{\infty} \hat{E}_{k,l} \hat{P}_{i,k-m-l} \tilde{r}_{i,m,n}^{(2)} \]

where we used the substitutions \( m + m' = k, n + n' = l \).

In the formalism of \((6q + 6)\)-dimensional matrices and vectors used in (63) the final result for the terms \( \tilde{r}_{i,m,n}^{(2)} \) and \( \tilde{r}_{i,m,n}^{(2)} \) is written as

\[ \sum_{k,l=-\infty}^{\infty} \hat{E}_{k,l} \hat{P}_{k-m,l-n} \tilde{r}_{i,m,n}^{(2)} \tilde{r}_{i,m,n}^{(2)} \]

where the diagonal matrix \( \hat{P}_{i,m-n} \) is given by (58) and the \((6q + 6)\)-dimensional vector \( \tilde{r}_{i,m,n}^{(2)} \) is defined by

\[ \tilde{r}_{i,m,n}^{(2)} = \begin{pmatrix} 0, 0, 0, \tilde{r}_{i,m,n}^{(2)} \tilde{r}_{i,m,n}^{(2)} \tilde{r}_{i,m,n}^{(2)} \tilde{r}_{i,m,n}^{(2)} \tilde{r}_{i,m,n}^{(2)} \end{pmatrix}^T. \]

The terms \( \tilde{r}_{i,m,n}^{(2)} \), which are proportional to the amplitudes \( A_{i,m,n} \) investigated in the previous section can be treated in a similar manner as the terms \( \tilde{r}_{i,m,n}^{(2)} \) and \( \tilde{r}_{i,m,n}^{(2)} \). From (34), (35), (42) and (43) it is easy to obtain the following expressions for the second order of perturbation

\[ \tilde{t}^{(2)}_{i,x,L} = \sum_{n,m=-\infty}^{\infty} \hat{E}_{m,n} \left[ f_{i,x} \left( A_{x,i,m,n} e^{\hat{\theta}_i} + A_{x,i,m,n} e^{-\hat{\theta}_i} \right) \right. \]

\[ \left. - A_{z,i-1,m,n} e^{\hat{\theta}_i} - A_{z,i-1,m,n} e^{-\hat{\theta}_i} \right] \]

\[ + i \hat{b}_{i,m,n} \left( A_{y,i,m,n} e^{\hat{\theta}_i} - A_{y,i,m,n} e^{-\hat{\theta}_i} \right) \]

\[ + i \hat{b}_{i-1,m,n} \left( A_{y,i-1,m,n} e^{\hat{\theta}_i} - A_{y,i-1,m,n} e^{-\hat{\theta}_i} \right) \]

\[ + i \hat{b}_{i-1,m,n} \left( A_{y,i-1,m,n} e^{\hat{\theta}_i} - A_{y,i-1,m,n} e^{-\hat{\theta}_i} \right) \]

\[ \right]. \]

(71)
\[ \sigma_{\beta}^{(2b)} = \sum_{m,n=-\infty}^{\infty} \hat{E}_{m,n} \left[ -f_{i,x} N \left( \hat{A}_{x,i,n} + \hat{A}_{y,i,n} \right) - \hat{A}_{x,i,n} \right] \]

\[ + \left. \hat{A}_{y,i,n} \right|_{m,n} \left( \hat{A}_{x,i,n} \right) \hat{A}_{y,i,n} \right|_{m,n} \left( \hat{A}_{x,i,n} \right) - \hat{A}_{x,i,n} \right] \]

\[ - \hat{A}_{x,i,n} \right|_{m,n} \left( \hat{A}_{x,i,n} \right) \hat{A}_{y,i,n} \right|_{m,n} \left( \hat{A}_{x,i,n} \right) - \hat{A}_{x,i,n} \right] \]

\[ \left( \hat{A}_{x,i,n} \right) \hat{A}_{y,i,n} \right|_{m,n} \left( \hat{A}_{x,i,n} \right) - \hat{A}_{x,i,n} \right] \]

The functions \( f_i \) and their derivatives \( f_{i,x} \) and \( f_{i,y} \) can be written using the Fourier components \( \hat{P}_{i,m,n} \) introduced in (3). The parts of the above expression that depend on the spatial coordinates \( x \) and \( y \) can be rewritten as

\[ \hat{E}_{m,n} \left[ f_{i,x} \right] = \sum_{m',n'=\infty}^{\infty} \hat{E}_{m+m',n+n'} \hat{P}_{j,m',n'} \left[ \frac{1}{iM'} \right] \]

\[ = \sum_{k,l=\infty}^{\infty} \hat{E}_{k,l} \hat{P}_{j,k-l,\infty} \left[ \frac{1}{i(K-M)} \right], \]

where we used the substitutions \( m+m' = k \) and \( n+n' = l \). The terms corresponding to \( \sigma_{\beta}^{(2b)} \) and \( \sigma_{\beta}^{(2b)} \) can be expressed in the matrix form as

\[ \sum_{k,l=\infty}^{\infty} i \hat{E}_{k,l} \hat{P}_{j,k-l,\infty} \hat{N}_{m,n,k,l} \hat{A}_{m,n} \]

where the (6q + 6)-dimensional square matrix \( \hat{N}_{m,n,k,l} \) is defined as

\[ \hat{N}_{m,n,k,l} = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & \cdots & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} \]

\[ \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & \cdots & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} \]

\[ \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & \cdots & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} \]

\[ \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & \cdots & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} \]

\[ \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & \cdots & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} \]

\[ \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & \cdots & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} \]

\[ \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & \cdots & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} \]

\[ \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & \cdots & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} \]

\[ \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & \cdots & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix} \]

With the help of the results presented above it is possible to write the boundary conditions (62) in matrix form.
If we combine the terms (63), (69) and (73) into one equation and remove the common factor, \( \sum_{k,l=\pm \infty} \hat{E}_{k,l} \) from this equation then we obtain the following result

\[
\hat{A}_{k,l}\beta^{(2)} = \sum_{m,n=-\infty}^{\infty} \hat{\mathcal{P}}_{k-m,l-n} \times \left[ \hat{\mathcal{P}}_{m-v,n}\hat{\mathcal{R}}_{m,n} - i\hat{\mathcal{N}}_{m,n,k,l}\hat{A}_{m,n} \right].
\]

(76)

In the previous section it was shown how can be the vector of amplitudes for the first order of perturbation \( \hat{A}_{m,n}^{(1)} \) calculated. After this vector is expressed from (53) and substituted into the Equation (76) the following formula for the vector of amplitudes corresponding to the second order of perturbation is obtained

\[
\hat{A}_{k,l}^{(2)} = \hat{A}_{k,l}^{(1)} \sum_{m,n=-\infty}^{\infty} \hat{\mathcal{P}}_{k-m,l-n} \times \left[ \hat{\mathcal{P}}_{m-v,n}\hat{\mathcal{R}}_{m,n} + \hat{\mathcal{N}}_{m,n,k,l}\hat{A}_{m,n}^{-1}\hat{\mathcal{P}}_{m-v,n}\hat{\mathcal{R}}_{m,n} \right].
\]

(77)

2.7. Amplitudes of reflected and transmitted light

In order to obtain quantities measurable by ellipsometry and spectrophotometry in specular reflection we must calculate the mean values of the amplitudes corresponding to averaging over different realizations of random roughness. Since the amplitudes \( \hat{A}_{m,n}^{(1)} \) corresponding to the first order of perturbation are linear in the coefficients \( \hat{\mathcal{P}}_{k,m,n} \) it is obvious that according to (4) the mean values of these amplitudes must vanish, i.e. \( \langle \hat{A}_{m,n}^{(1)} \rangle = 0 \). The situation is more complicated for the mean values of amplitudes \( \hat{A}_{k,l}^{(2)} \) corresponding to the second order of perturbation. These amplitudes exhibit quadratic dependence on the coefficients \( \hat{\mathcal{P}}_{k,m,n} \), therefore, their mean values must be calculated using the rules described in (4), (5). The terms quadratic in \( \hat{\mathcal{P}}_{k,m,n} \) in Equation (77) can be written as \( \hat{\mathcal{P}}_{m',n'}\hat{\mathcal{X}}\hat{\mathcal{P}}_{k',l'} \) where the indices are given as \( m' = k - m, n' = l - n, k' = m - v, l' = n \) and the \( (6q + 6) \times (6q + 6) \) matrix \( \mathcal{X} \) is either the unit matrix or it is equal to \( \hat{\mathcal{N}}_{m,n,k,l}\hat{A}_{m,n}^{-1} \). From (4) it follows that the mean value of this expression vanishes unless the indices satisfy the conditions \( m' \neq -k' \) and \( n' \neq -l' \), i.e.

\[
\langle \hat{\mathcal{P}}_{m',n'}\mathcal{X}\hat{\mathcal{P}}_{k',l'} \rangle_{(m',n') \neq (-k',-l')} = 0.
\]

(78)

This means that the mean value of the amplitudes \( \hat{A}_{k,l}^{(2)} \) is zero for \( k \neq v \) or \( l \neq 0 \). This condition says that the second order of perturbation gives nonzero corrections only to waves propagating in directions corresponding to coherent light. For \( m' = -k', n' = -l' \) the mean values can be expressed as

\[
\langle \hat{\mathcal{P}}_{m',n'}\mathcal{X}\hat{\mathcal{P}}_{-m',-n'} \rangle = a^2 \hat{\mathcal{W}}_{m',n'} \circ \mathcal{X},
\]

(79)

where \( \circ \) denotes the Hadamard product. The Hadamard product of two matrices \( \mathcal{X} \) and \( \mathcal{Y} \) with the same dimension \( (\mathcal{X} \circ \mathcal{Y})_{ij} = X_{ij}Y_{ij} \). The \( (6q + 6) \times (6q + 6) \) matrix \( \hat{\mathcal{W}}_{m',n'} \) in (79) is defined as

\[
\begin{pmatrix}
\hat{W}_{1,1} & \hat{W}_{1,2} & \hat{W}_{1,3} & \cdots & \hat{W}_{1,q} \\
\hat{W}_{2,1} & \hat{W}_{2,2} & \hat{W}_{2,3} & \cdots & \hat{W}_{2,q} \\
\hat{W}_{3,1} & \hat{W}_{3,2} & \hat{W}_{3,3} & \cdots & \hat{W}_{3,q} \\
\vdots & \vdots & \vdots & \ddots & \vdots \\
\hat{W}_{q,1} & \hat{W}_{q,2} & \hat{W}_{q,3} & \cdots & \hat{W}_{q,q}
\end{pmatrix}_{m',n'}.
\]

(80)

with the \( 6 \times 6 \) matrices \( \hat{W}_{i,j,m',n'} \) defined by means of the PSDF \( \hat{\mathcal{W}}_{i,j,m',n'} \) (5) as

\[
\hat{W}_{i,j,m',n'} =
\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 \\
0 & \hat{W}_{i,j} & \hat{W}_{i,j} & \hat{W}_{i,j} & \hat{W}_{i,j} & 0 \\
0 & \hat{W}_{i,j} & \hat{W}_{i,j} & \hat{W}_{i,j} & \hat{W}_{i,j} & 0 \\
0 & \hat{W}_{i,j} & \hat{W}_{i,j} & \hat{W}_{i,j} & \hat{W}_{i,j} & 0 \\
0 & \hat{W}_{i,j} & \hat{W}_{i,j} & \hat{W}_{i,j} & \hat{W}_{i,j} & 0 \\
0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}_{m',n'}.
\]

(81)

The indices \( m',n' \) used as subscripts to the above matrices are used to indicate that the elements of these matrices also depend on \( m',n' \). By using the formula (79) it is possible to express the mean value of (77) in the case \( k = v, l = 0 \). As was discussed above, this is the only case in which this mean value is nonzero. The result can be written in a closed form as

\[
\begin{pmatrix}
\hat{A}_{k,l}^{(2)} \\
\hat{A}_{k,l}^{(2)} \\
\hat{A}_{k,l}^{(2)} \\
\hat{A}_{k,l}^{(2)}
\end{pmatrix}_{v,0} = a^2 \hat{\mathcal{X}}_{k,l}^{-1} \sum_{m,n=-\infty}^{\infty} \left[ \hat{\mathcal{W}}_{m,n} - m,n \circ \mathcal{X} \hat{\mathcal{R}}_{m,n} \right].
\]

(82)

So far we have assumed that the problem that we are trying to solve is periodic in the \( x - y \) plane with period \( L \). This periodicity resulted in the fact that the spatial frequencies \( M \) and \( N \) can take only discrete values with the distance between the neighbouring values determined by the parameter \( a = 2\pi/L \). This limitation can be removed if we perform the limit \( L \to \infty \) in which the value \( a \) goes to zero, thus, the spectrum of spatial frequencies becomes continuous. In this limit the sums in (82) change into integrals and the dependence on the discrete indices \( m, n \)
The optical constants of silicon single crystal are taken from (49), the optical constants of silicon dioxide are taken from (50) and the optical constant of silicon nitride are taken from (51).

The random roughness of the boundaries is described by the PSDF given as the Gaussian function

$$
\hat{W}_{i,j,m,n} = \frac{c_{ij}\sigma_i\tau^2}{4\pi} e^{-\frac{\tau^2}{(M^2+N^2)}},
$$

where the symbol $\sigma_i$ denotes the rms value of heights of roughness of the $i$th boundary and the symbol $\tau$ is the correlation length. Note that the correlation length is assumed to be identical for all boundaries. For $i = j$ the coefficients $c_{ij}$ are equal to unity, i.e. $c_{ij} = 1$. In this section we will present results for two models of rough boundaries:

- The first model assumes independently rough boundaries. In this model, there is no correlation among roughnesses of different boundaries, thus, all coefficients are zero ($c_{ij} = 0$) for $i \neq j$. The rms values of heights will be chosen to be the same for all boundaries $\sigma_i = \sigma$.
- The second model assumes identically rough boundaries. In this model the roughness of all boundaries is identical from both the geometrical and statistical points of view. Therefore, the rms values of heights are identical $\sigma_i = \sigma$ and all coefficients $c_{ij}$ are equal to unity, i.e. $c_{ij} = 1$.

Apart from the results corresponding to the above two models of roughness we will present also two other results corresponding to:

- The approximate method called the IRA based on the assumption that the individual rough boundaries are considered to be isolated (published in (25)). The rms values of heights will be chosen to be the same for all boundaries $\sigma_i = \sigma$.
- The system of layers with smooth boundaries (i.e. without roughness). These results are presented in order to show the influence of boundary roughness on the reflectometric and ellipsometric data.

The calculated spectral dependencies of reflectance and associated ellipsometric parameters for thicknesses of layers 75 nm/60 nm/75 nm, the rms value of heights $\sigma = 7$ nm and the correlation length $\tau = 70$ nm are introduced in Figure 3. The differences between results corresponding to different models with rough boundaries are small compared to differences from the results obtained for the model with smooth boundaries. The differences are higher for higher photon energies where the ratio $\sigma/\lambda_0$ is larger.

3. Numerical examples

In this section, the numerical simulations are used to study the influence of randomly rough boundaries on measurable optical quantities. The following three-layer system is selected for this numerical simulation: silicon oxide (SiO$_2$)/silicon nitride (Si$_3$N$_4$)/silicon oxide (SiO$_2$) on semi-infinite silicon single crystal substrate. The influence of randomly rough boundaries will be studied by means of simulated spectral dependencies of reflectance for normal angle of incidence $\theta_0 = 0^\circ$ and associated ellipsometric parameters corresponding to reflected light for angle of incidence $\theta_0 = 75^\circ$. The reflectance is calculated from the amplitudes (84) as

$$
R = |\hat{r}_0^s|^2 = |\hat{r}_0^p|^2.
$$

The associated ellipsometric parameters are calculated as (3)

$$
I_s = -\frac{\hat{r}_0^p r_0^s + \hat{r}_0^s r_0^p}{|\hat{r}_0^s|^2 + |\hat{r}_0^p|^2},
I_c = -\frac{\hat{r}_0^p r_0^s + \hat{r}_0^s r_0^p}{|\hat{r}_0^s|^2 + |\hat{r}_0^p|^2},
I_n = \frac{|\hat{r}_0^s|^2 - |\hat{r}_0^p|^2}{|\hat{r}_0^s|^2 + |\hat{r}_0^p|^2}.
$$

The optical constants of silicon single crystal are taken from (49), the optical constants of silicon dioxide are taken from (50) and the optical constant of silicon nitride are taken from (51).

The random roughness of the boundaries is described by the PSDF given as the Gaussian function

$$
\hat{W}_{i,j,m,n} = \frac{c_{ij}\sigma_i\tau^2}{4\pi} e^{-\frac{\tau^2}{(M^2+N^2)}},
$$

where the symbol $\sigma_i$ denotes the rms value of heights of roughness of the $i$th boundary and the symbol $\tau$ is the correlation length. Note that the correlation length is assumed to be identical for all boundaries. For $i = j$ the coefficients $c_{ij}$ are equal to unity, i.e. $c_{ij} = 1$. In this section we will present results for two models of rough boundaries:

- The first model assumes independently rough boundaries. In this model, there is no correlation among roughnesses of different boundaries, thus, all coefficients are zero ($c_{ij} = 0$) for $i \neq j$. The rms values of heights will be chosen to be the same for all boundaries $\sigma_i = \sigma$.
- The second model assumes identically rough boundaries. In this model the roughness of all boundaries is identical from both the geometrical and statistical points of view. Therefore, the rms values of heights are identical $\sigma_i = \sigma$ and all coefficients $c_{ij}$ are equal to unity, i.e. $c_{ij} = 1$.

Apart from the results corresponding to the above two models of roughness we will present also two other results corresponding to:

- The approximate method called the IRA based on the assumption that the individual rough boundaries are considered to be isolated (published in (25)). The rms values of heights will be chosen to be the same for all boundaries $\sigma_i = \sigma$.
- The system of layers with smooth boundaries (i.e. without roughness). These results are presented in order to show the influence of boundary roughness on the reflectometric and ellipsometric data.

The calculated spectral dependencies of reflectance and associated ellipsometric parameters for thicknesses of layers 75 nm/60 nm/75 nm, the rms value of heights $\sigma = 7$ nm and the correlation length $\tau = 70$ nm are introduced in Figure 3. The differences between results corresponding to different models with rough boundaries are small compared to differences from the results obtained for the model with smooth boundaries. The differences are higher for higher photon energies where the ratio $\sigma/\lambda_0$ is larger.
Figure 3. The spectral dependencies of the reflectance $R$ and associated ellipsometric parameters $I_s$, $I_c$, $I_n$. The thicknesses of the layers are 75 nm/60 nm/75 nm, the values of the roughness parameters are $\sigma = 7$ nm and $\tau = 70$ nm.

Figure 3 depicts differences between individual models of roughness for one combination of thicknesses of layers and values of $\sigma$ and $\tau$. We are going to show how these differences change if the thicknesses or values of $\sigma$ and $\tau$ change. For this purpose it is convenient to introduce the rms values of differences from the results corresponding to the model with independently rough boundaries.

Figure 4. The dependence of the rms values of differences $\delta_R$ and $\delta_I$ on the total thickness $h$ of the three-layer system, rms values of heights $\sigma$ with fixed ratio of $\sigma/\tau$ and correlation length $\tau$ with fixed $\sigma$.

These quantities are defined as

$$
\delta_R = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (R_i^a - R_i^b)^2},
$$

$$
\delta_I = \sqrt{\frac{1}{3N} \sum_{i=1}^{N} \left( (I_s^a - I_s^b)^2 + (I_c^a - I_c^b)^2 + (I_n^a - I_n^b)^2 \right)},
$$

(87)

where the reflectance $R_i^a$ and the ellipsometric parameters $I_{s,i}^a$, $I_{c,i}^a$, $I_{n,i}^a$ calculated for different photon energies are distinguished by the lower index $i$. The upper indices $a$ and $b$ are used to distinguish the reflectances and ellipsometric parameters corresponding to different models of rough boundaries, i.e. between model with identically
rough boundaries, independently rough boundaries and the IRA model. The values inside sums in (87) are calculated for \( N = 531 \) equidistant points in the spectral range 1.2–6.5 eV (191–1033 nm).

The dependencies of the differences \( \delta_R \) and \( \delta_I \) on the thicknesses of the layers, the rms value of heights \( \sigma \) and the correlation length \( \tau \) are depicted in Figure 4. The top figure depicts the dependence on the total thickness of the three-layer system with roughness parameters chosen as \( \sigma = 7 \text{ nm} \) and \( \tau = 70 \text{ nm} \). The ratio of thicknesses is kept constant at 5:4:5, i.e. the thicknesses of the layers change from 18.25 nm/15 nm/18.25 nm to 300 nm/240 nm/300 nm. From this figure, it is evident that the influence of the correlation between the rough boundaries decreases with increasing thickness of the layers. This implies that there are the thickness values above which the IRA represents a sufficient approximation. The middle figure shows the dependence on the rms values of heights \( \sigma \) with thicknesses of layers chosen as 75 nm/60 nm/75 nm. The correlation length is chosen such that the ratio \( \sigma/\tau = 1/10 \) is kept constant. This means that the rms value of slopes of the roughness is kept at constant value. The differences \( \delta_R \) and \( \delta_I \) are growing approximately quadratically with the increasing rms value of heights. This result is expectable because the corrections to the electric filed amplitudes are proportional to the PSDF (83) which is quadratic in \( \sigma \) (86). The bottom figure depicts the dependence on the correlation length \( \tau \). The thicknesses of the layers are again 75 nm/60 nm/75 nm and the rms value of heights is kept at \( \sigma = 7 \text{ nm} \). From this figure it seems that the dependence of the differences \( \delta_R \) and \( \delta_I \) on the correlation length \( \tau \) is approximately linear. The values for the ratio \( \sigma/\tau \) greater than 1/5 (the values of \( \tau \) less than 35 nm) were not calculated since the RRT is valid only if the slopes of the roughness are small.

4. Conclusion

In this paper, the second-order RRT is employed for calculating the spectral dependencies of reflectance and associated ellipsometric parameters of the multi-layer systems with boundaries exhibiting slight random roughness. The presented method takes into account the propagation of the perturbed electromagnetic fields (waves) occurring inside the individual randomly rough layers. This means that the obtained results are exact up to the second order of perturbation. It is necessary to point out that the restriction to the second order of perturbation represents only one approximation in our calculations, i.e. the propagation of the perturbed waves in the multi-layer system is solved exactly. This approach represents the improvement in comparison with the approximate approach presented in paper (25) in which the propagation of the perturbed waves is neglected. Another benefit of the exact approach presented in this paper is that it is possible to consider statistical correlations among boundary roughnesses. This is something that has not been done before for the multi-layer system. The disadvantage of the presented method is that it requires calculation of inverse matrices. The sizes of these matrices grow with the number of layers, therefore, the numerical calculations are slow if the number of layers is large.

The derived formulae for normal reflectance and associated ellipsometric parameters are used to perform calculations for numerical examples of the selected three-layer system. This system consists of three layers of \( \text{SiO}_2/\text{Si}_3\text{N}_4/\text{SiO}_2 \) on top of Si substrate. The purpose of these numerical examples is to show the influence of the individual geometrical parameters, i.e. the total thickness of the layered system and roughness parameters, on the mentioned optical quantities.

The emphasis is placed on the comparison of the numerical results obtained for the models with statistically identical and independently randomly rough boundaries. It is shown that the differences among optical quantities corresponding to these models grow approximately quadratically with the rms value of heights which is expectable result. The differences also seem to grow approximately linearly with the correlation length. The numerical results also indicate that the influence of the statistical correlation among randomly rough boundaries is larger for smaller thicknesses of the layers. The results of the models with statistically independently randomly rough boundaries and identical rough boundaries are also compared with results obtained by the approximate method presented in (25). For small thicknesses of layers the approximate method shows large differences from the correctly calculated values, therefore, one can conclude that in this case the propagation of perturbed waves in the multi-layer systems cannot be neglected. Note that for single layer the limitations of the approximate method were discussed in detail in (25). In conclusion it should be pointed out that the exact approach of the RRT presented here is also important for applied research and optics industry applications because the multi-layer systems with slightly rough boundaries are frequently encountered in these fields. The formulae of this exact approach enable us to improve the optical characterization of these rough multi-layer systems.

Disclosure statement

No potential conflict of interest was reported by the authors.
Funding

This work was supported by the project LO1411 (NPUI) funded by Ministry of Education, Youth and Sports of the Czech Republic.

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