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DETERMINATION OF OPTICAL CONSTANTS OF VERY THIN FILMS: AN EXACT ANALYTICAL APPROACH

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Inverse optical problems are of major importance for many scientific and engineering projects. To name a few, solutions of nonlinear equations are needed for the determination of optical performance of stable or light sensitive thin films. In this communication we briefly describe an analytical method for finding the roots of a set of nonlinear equations by transformation of the equation system into a multivariant polynomial form. The main advantage of the method is that all possible solutions are found without any initial guess of the unknown parameters. The inverse optical problem of determination of complex refractive index and physical thickness of a thin film from spectrophotometric experimental data is discussed.

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1. Introduction

The mathematical solutions of inverse problems in modern optics consist in determination of the roots of a system of nonlinear equations [1,2]. Powerful methods for numeric solutions are developed [1], but they are strongly dependent on the choice of the initial guess of the unknown parameters. Besides, they have the inconvenience of having multiple solutions, related to local minima in the solution process, or – even worse – lack of solutions. These problems are well known and they make very difficult the choice of the optimal (by some criteria) scientific or engineering solution.

In this communication we propose an exact analytical method for inverse problem so lutions that renders possible, for a certain class of optical problems, the determination of the global minima in the parameter space. A similar method is tried and successfully implemented for the exact simultaneous correction of several aberrations in the optical system design [3]. Here, an example is given that will illustrate the potentialities of the method. It is related to the determination of the complex refractive index ($\tilde{n}$) and physical thickness (d) of very thin films, stable or with photoinduced changes.

2. Transformation of the system of nonlinear equations and successive elimination of the unknowns:

Let us consider a system of q nonlinear equations with q unknown parameters x_j $(j = 1,\ldots,q)$. In order to find the exact solutions of the system we transform each equation in a mixed polynomial as follows:

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$$
\begin{aligned}
&a_l(x_2,...,x_q)x_1^l + a_{l-1}(x_2,...,x_q)x_1^{l-1} + ... + a_1(x_2,...,x_q)x + a_0(x_2,...,x_q) = 0\,;\\
&b_m(x_2,...,x_q)x_1^m + b_{m-1}(x_2,...,x_q)x_1^{m-1} + ... + b_1(x_2,...,x_q)x + b_0(x_2,...,x_q) = 0\,;\\
&c_n(x_2,...,x_q)x_1^n + c_{n-1}(x_2,...,x_q)x_1^{n-1} + ... + c_1(x_2,...,x_q)x + c_0(x_2,...,x_q) = 0\,;\\
&\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\\
&g_p(x_2,...,x_q)x_1^p + g_{p-1}(x_2,...,x_q)x_1^{p-1} + ... + g_1(x_2,...,x_q)x + g_0(x_2,...,x_q) = 0\,.
\end{aligned}
\tag{2.1}
$$

The coefficients {*a, b, c,…,g*} are functions of all unknown variables but x_1. The coefficients *l, m, n,…,p* are equal to the highest power of the parameter x_1 and correspond to the highest index of $a_l, b_m, c_n,\ldots,g_p$.

The elimination of one of the unknown parameters is based on a modified approach developed by Viete [4]. The idea is to transform the system of *q* nonlinear equation in a system of (*q - 1*) equations. We make use of the resultant (or eliminant) determinants of the first equation together with the rest of the system equations. For the firs two equation of (2.1) having coefficients a_l and b_m the eliminant determinant D_{ab} is [5]:

$$
D_{ab} = \begin{vmatrix}
a_l & a_{l-1} & . & . & . & a_0 & 0 & . & . & . & 0\\
0 & a_l & a_{l-1} & . & . & . & a_0 & 0 & 0 & 0 & 0\\
. & . & . & . & . & . & . & . & . & . & .\\
0 & . & . & . & 0 & a_l & a_{l-1} & . & . & . & a_0\\
b_m & b_{m-1} & . & . & . & b_0 & 0 & . & . & . & 0\\
0 & b_m & b_{m-1} & . & . & . & b_0 & 0 & 0 & 0 & 0\\
. & . & . & . & . & . & . & . & . & . & .\\
0 & . & . & . & 0 & b_m & b_{m-1} & . & . & . & b_0
\end{vmatrix}.
\tag{2.2}
$$

D_{ab} has (l + m) columns and (m + l) rows. The number of rows, constructed with the '*a*' coefficients is equal to *m* and the number of rows with '*b*' coefficients is equal to *l*. We present the D_{ab} determinant as a polynomial of powers of x_2:

$$
D_{ab} = D_s(x_3,...,x_q)x_2^s + D_{s-1}(x_3,...,x_q)x_2^{s-1} + ... + D_1(x_3,...,x_q)x_2 + D_0(x_3,...,x_q),
\tag{2.2a}
$$

where *s* is the highest power of x_2, and $D_s,...,D_0$ are functions of all unknown variables but x_1 and x_2 .

Thus the system of *q* equations with *q* unknowns (2.1) is substituted with a system of (*q - 1*) equations with (*q - 1*) unknowns.

$$
D_{ab} = 0;\ D_{ac} = 0;\ldots;\ D_{ag} = 0.
\tag{2.3}
$$

By analogy with the elimination of x_1 we lower the number of equations to (*q - 2*) with *(q - 2)* unknowns, etc till we obtain one polynomial with one unknown x_q. If we find all the roots of this polynomial equation, we can find all possible solutions of the initial system (2.1).

3. Determination of (*ñ*, *d*) of very thin films

The method described above is applied to solve the system of three equations (3.1) that relate the optical response of a very thin film (the film front side reflectance (*R*), the film back side reflectance (*R´*) and the transmittance (*T*)) with the unknown quantities - the physical thickness, the real (ε_1) and imaginary (ε_2) part of complex permitivitty ($\varepsilon = \varepsilon_1 - i\varepsilon_2$) of the film. Utilizing the nanothickness of the film we develop the trigonometric functions in thin film characteristic matrix elements to the third order in terms of $\tilde{n}d/\lambda$ ($\tilde{n} = \sqrt{\varepsilon}$, λ is the wavelength) and derive approximated expression for R, R' and T. Forming the expressions $(1 + R)/T$, $(1 - R)/T$, $(1 - R')/T$, which are simpler then those for R, R', T, we obtain the following system:

$$\frac{1+R}{T}=\frac{(\varepsilon_2 n_s - n_s\varepsilon_2\varepsilon_1)\omega^3 d^3}{3n_s}+\frac{[(1-\varepsilon_1)(n_s{}^2-\varepsilon_1)+\varepsilon_2{}^2]\omega^2 d^2}{2n_s}+\frac{2\omega d\varepsilon_2 n_s+(n_s{}^2+1)}{2n_s};$$

$$\frac{1-R}{T}=\frac{(\varepsilon_2 n_s{}^2-\varepsilon_2\varepsilon_1)\omega^3 d^3}{3n_s}+\frac{\omega d\varepsilon_2+n_s}{n_s}; \tag{3.1}$$

$$\frac{1-R'}{T}=\frac{(\varepsilon_2-\varepsilon_2\varepsilon_1)\omega^3 d^3}{3}+\omega d\varepsilon_2+1,$$

where $\omega = 2\pi/\lambda$ is the wave number, n_s is a refractive index of the substrate.

We perform algebraic transformations and substitutions in the system (3.1) to make easier the application of the method. First we introduce a new variable: $V=\omega d$ and the constants: $p_1=\frac{3n_s(1-R)}{T}$, $p_2=\frac{3(1-R')}{T}$, $p_3=\frac{6n_s(1+R)}{T}$. After this we subtract the third equation of (3.1) from the second one and obtain the equation:

$$V^3\varepsilon_2 - L = 0, \tag{3.2}$$

where $L=\frac{3(n_s-1)+p_2-p_1}{1-n_s^2}$, then we substitute (3.2) in the second equation of (3.1) and derive the equation:

$$V^2\varepsilon_1 - V^2 M - 3 = 0, \tag{3.3}$$

where $M = (Ln_s^2 - p_1 + 3n_s)/L$.

Thus the initial system (3.1) is converted into set of three polynomials:

$$a_1\varepsilon_1 + a_0 = 0; \tag{3.4a}$$

$$b_1\varepsilon_2 + b_0 = 0; \tag{3.4b}$$

$$c_2\varepsilon_2^2 + c_1\varepsilon_2 + c_0 = 0, \tag{3.4c}$$

where $a_1 = V^2$, $a_0 = -3 - MV^2$, $b_1 = V^3$, $b_0 = -L$, $c_2 = 3V^2$, $c_1 = 2n_sV^3(1 - \varepsilon_1) + 6Vn_s$, $c_0 = 3V^2\varepsilon_1^2 - 3V^2\varepsilon_1(1 + n_s^2) + 3V^2n_s^2 + 3(n_s^2 + 1) - p3$.

The method of excluding the unknown quantities is applied to solve the system (3.4). First we exclude ε_2, using equations (3.4b) and (3.4c). The eliminant determinant D_{bc} of the equations (3.4b) and (3.4c) is:

$$D_{bc}=\begin{vmatrix} b_1 & b_0 & 0 \\ 0 & b_1 & b_0 \\ c_2 & c_1 & c_0 \end{vmatrix}.$$

Solving $D_{bc} = 0$, we obtain a polynomial for ε_1:

$$e_2\varepsilon_1^2 + e_1\varepsilon_1 + e_0 = 0, \tag{3.5}$$

where $e_2 = 3V^6$, $e_1 = -V^4[2Ln_s + 3V^2(1+n_s^2)]$ and $e_0 = 3n_s^2V^6 + V^4[3(n_s^2 + 1) + 2Ln_s - p_3] + 6Ln_sV^2 + 3L^2$.

The second step is to exclude ε_1. With the purpose of this, (3.5) is used together with equations (3.4a). The eliminant determinant D_{ae} of the equations (3.5) and (3.4a) is:

$$D_{ae} = \begin{vmatrix} a_1 & a_0 & 0 \\ 0 & a_1 & a_0 \\ e_2 & e_1 & e_0 \end{vmatrix}.$$

Solving $D_{ae} = 0$, we derive a polynomial of third degree for $U = V^2 = (\omega d)^2$:

$$f_3U^3 + f_2U^2 + f_1U + f_0 = 0, \tag{3.6}$$

where $f_3 = 3(n_s^2 - M)(1 - M)$, $f_2 = -p_3 - 6(n_s^2 + 1) + 2(-MLn_s + Ln_s + 9M)$, $f_1 = 27$ and $f_0 = 3L^2$. In fact this is polynomial for d. Now we can find all roots of the polynomial (3.6) for U and all roots of d. When d is substituted in (3.4b) and (3.4a) ε_2 and ε_1 are obtained, respectively. As a verification of the accuracy of the obtained solutions, we substitute each triad (ε_1, ε_2, d) in the equations (3.1) and obtain results quite close to computer epsilon (zero).

To illustrate the application of the described method, we made realistic numeric simulation of spectrophotometric measurements of nano-film, deposited on transparent substrate, in the 450 – 1500 nm spectral range. A film with values of ε_1 and ε_2 typical for amorphous semiconductors (As-S, As-Se, Ge-Se, etc.) and thickness of 15 nm is considered. A semi-infinite substrate with ε_s very closed to the values of BK7 glass is assumed. The spectral dependence of model $\varepsilon_1(\lambda)$, $\varepsilon_2(\lambda)$ and $\varepsilon_s(\lambda)$ are presented in Fig. 1.

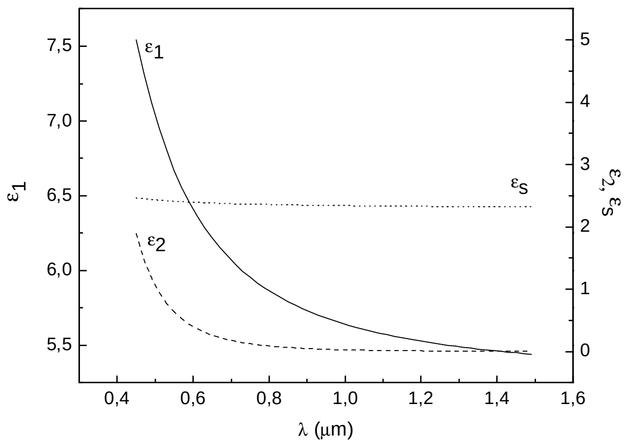

Fig. 1. Spectral dependence of model $\varepsilon_1(\lambda)$(—), $\varepsilon_2(\lambda)$(---) and $\varepsilon_s(\lambda)$(…).

For the needs of the inverse problem, we first evaluate at each wavelength (R, R', T) by the help of procedure reported in [6], and then we calculated (ε_1, ε_2, d), using the above described procedure.

The polynomial (3.6), which is cubic for U, has three real roots for each wavelength – two positive and one negative. The thickness d has 4 real and 2 complex roots, but only 2 positive roots have physical meaning. The dispersions of those roots (d_1 and d_2) are presented in Fig. 2.

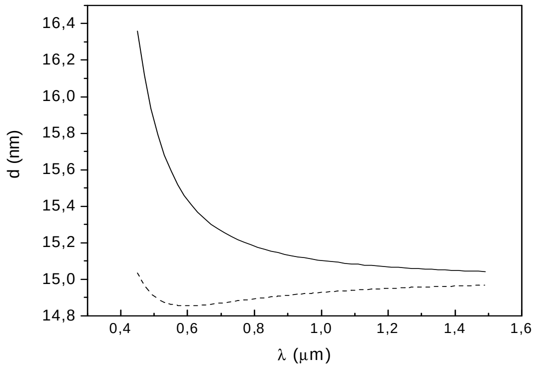

Fig. 2. Spectral dependence of the two real roots: d_1 (—) and d_2 (---) of the polynomial (3.6).

With d_1 and d_2 we solve the equation (3.4a) for ε_1 and equation (3.4b) for ε_2.
As a measure of the accuracy of the solutions we use the relative uncertainties:

$$\frac{\Delta\varepsilon_1}{\varepsilon_1} = \frac{\varepsilon_1(\lambda) - \varepsilon_1{}^*(\lambda)}{\varepsilon_1(\lambda)}; \quad \frac{\Delta\varepsilon_2}{\varepsilon_2} = \frac{\varepsilon_2(\lambda) - \varepsilon_2{}^*(\lambda)}{\varepsilon_2(\lambda)} \quad ; \quad \frac{\Delta d}{d} = \frac{\mathrm{d}(\lambda) - \mathrm{d}^*(\lambda)}{\mathrm{d}(\lambda)},$$

where (ε_1, ε_2, d) are the model complex refractive index parameters and thickness and (ε_1^*, ε_2^*, d^*) are the estimated values. The results on spectral dependence of $\Delta\varepsilon_1/\varepsilon_1$, $\Delta\varepsilon_2/\varepsilon_2$ and $\Delta d/d$ for (ε_1, ε_2), calculated with two real values d_1 and d_2, are presented in Fig. 3a and 3b, respectively.

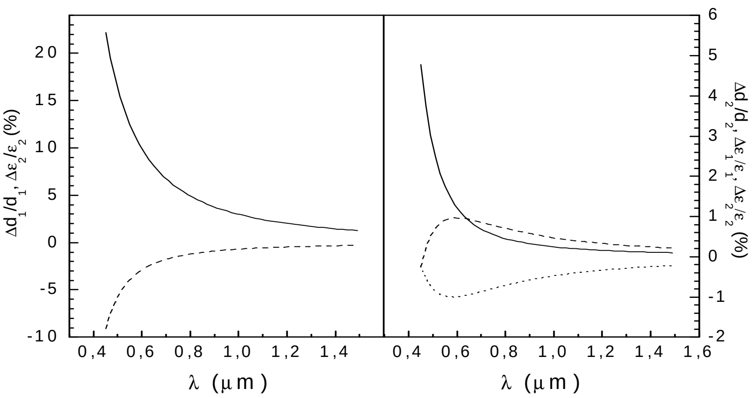

Fig. 3a. Spectral dependence of $\Delta d_1/d_1$(---) and $\Delta\varepsilon_2/\varepsilon_2$(—).

Fig. 3b. Spectral dependence of $\Delta d_2/d_2$(---), $\Delta\varepsilon_1/\varepsilon_1$(—) and $\Delta\varepsilon_2/\varepsilon_2$(…).

The (ε_1^*, ε_2^*) calculated with d_2, are closer to the model values: the maximum value of $\Delta\varepsilon_1/\varepsilon_1$ is ~5%, of $\Delta\varepsilon_2/\varepsilon_2$ is ~ -1% and of $\Delta d/d$ is ~1%. The values of ε_1^* calculated with d_1 are negative and that is why the dispersion of $\Delta\varepsilon_1/\varepsilon_1$ is not presented in Fig. 3a. We have to point out that the relatively high values of ($\Delta\varepsilon_1/\varepsilon_1$, $\Delta\varepsilon_2/\varepsilon_2$, $\Delta d/d$) are not because of inaccuracies in the inverse optical problem solutions but is due to the fact that equations (3.1) are approximate.

The absolute differences $\Delta T = T - T_{cal}$, $\Delta R = R - R_{cal}$ and $\Delta R' = R' - R'_{cal}$ (where T_{cal} is the transmittance, R_{cal} is front side reflectance and is R'_{cal} back side reflectance, calculated with the help of the triad (ε_1^*, ε_2^*, d_2^*) and exact matrix elements) can be also used as a measure for accuracy of the solutions. The results are presented in Fig. 4. ΔT has maximum value 1%, ΔR - 1.2% and $\Delta R'$-

1.4%. The aim of the future work is to decrease these differences, by developing the system (3.1), using the expansion of the matrix elements up to fourth order in terms of $\tilde{n}d/\lambda$.

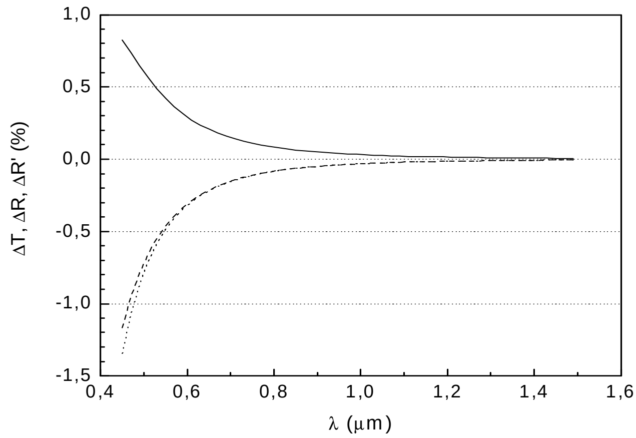

Fig. 4. Spectral dependence of ΔT(—), ΔR(---) and $\Delta R'$(…), where T_{cal}, R_{cal} and R'_{cal} are calculated, using (ε_1^*, ε_2^*, d_2^*).

4. Conclusion

We have presented an exact method for finding the solutions of a nonlinear system of equations of polynomial type. An example illustrates the potentialities of the method. The determination of the optical constants of a thin film with thickness of 15 nm shows that the different roots of the nonlinear system can be separated and the root with physical significance easily located.

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