JOURNAL OF OPTOELECTRONICS AND ADVANCED MATERIALS Vol. 9, No. 1, January 2007, p. 217 - 220

Analytical solution of the inverse optical problem for very thin films

B. HRISTOV, P. GUSHTEROVA*, P. SHARLANDJIEV
Central Laboratory for Optical Storage and Processing of Information, Bulgarian Academy of Sciences, "Acad. G. Bontchev" Str., Bl. 101, 1113 Sofia, Bulgaria.

The determination of the complex refractive index (*ñ*) and the physical thickness (*d*) of very thin films ($d \cong \lambda/50$, λ is the wavelength in VIS and NIR) is still a challenging task in the field of the inverse optical problems. The physical reality of these films makes difficult the application of methods, commonly used for the determination of *ñ* and *d* of thicker films. For several years we have been working on the development and the implementation of a method, designed especially for determination of *ñ* and *d* of very thin films. The nanothickness of the films allows us to develop in series of *ñd/λ* the Abelès characteristic matrix elements. Thus, approximated expressions for the film transmittance (T_f), front side (R_f) and backside (R'_f) reflectance are derived. For estimation of *ñ* and *d* we use the system $(1+R_f)/T_f$, $(1-R_f)/T_f$ and $(1-R'_f)/T_f$. Here we apply an exact analytical approach to solve the system, obtained by development of the Abelès characteristic matrix elements to the 4-th power. For the first time, the set of equations for $(1+R_f)/T_f$, $(1-R_f)/T_f$ and $(1-R'_f)/T_f$ is solved analytically. Besides the increase of the accuracy of the solutions, no initial information about the unknown parameters is needed.

(Received November 1, 2006; accepted December 21, 2006)

Keywords: Very thin films, Optical constants, Exact analytical approach

1. Introduction

The determination of the thin film optical constants (complex refractive index (*ñ*) and physical thickness, (*d*)), from experimental data on the film transmittance (T_f), front reflectance (R_f) and back reflectance (R'_f) is still a challenging problem. Due to the intensive development of nanotechnologies, the interest of the optical constants determination of thin homogeneous and composite films is revived. Wolter and Abeles [1,2] developed approximate methods for the determination of *ñ* or (*ñ*, *d*) from spectrophotometric measurements for thin metallic films. They used a limited development of the expressions for R_f, R'_f and T_f in orders of $(\tilde{n}d/\lambda)$, where λ is the wavelength.

We have developed a method [3,4,5] for the determination of $\tilde{n} = n - ik$ and *d* of very thin films, where *n* is the refractive index and *k* the extinction coefficient. General expressions for T_f, R_f and R'_f are derived by a development of the Abelès characteristic matrix elements up to a certain power of $(\tilde{n}d/\lambda)$. The system $(1+R_f)/T_f$, $(1-R_f)/T_f$ and $(1-R'_f)/T_f$ is readily used, because of its simplicity. The derived set of equations can be solved analytically or numerically. Recently, we apply an exact analytical approach to solve the system, obtained by development of the Abelès characteristic matrix elements up to a third power [4]. The equations of the system are transformed into a multivariant polynomial form. The main advantage of the exact analytical approach is that all possible solutions are found without any initial guess of the unknown parameters.

In this paper, we use the exact analytical approach to solve the system obtained, by developing the Abelès characteristic matrix elements up to the 4-th order. An example is given that illustrates the potentialities of the method.

2. Application of the exact analytical approach for determination of (*ñ, d*)

The initial system was derived elsewhere and has the form [3]:

$$\frac{1+R_f}{T_f} = -\frac{(0.5\varepsilon_2^2 + \varepsilon_1^2 - \varepsilon_2^2\varepsilon_1 + 0.5\varepsilon_2^2 n_s^2 - \varepsilon_1 n_s^2 + \varepsilon_1^2 n_s^2 - \varepsilon_1^3)\omega^4 d^4}{6n_s} + \frac{(\varepsilon_2 n_s - n_s\varepsilon_2\varepsilon_1)\omega^3 d^3}{3n_s} + \frac{[(1-\varepsilon_1)(n_s^2 - \varepsilon_1) + \varepsilon_2^2]\omega^2 d^2}{2n_s} + \frac{2\omega d\varepsilon_2 n_s + (n_s^2+1)}{2n_s}; \quad (1a)$$

$$\frac{1-R_f}{T_f} = \frac{\varepsilon_2^2\omega^4 d^4}{6} + \frac{(\varepsilon_2 n_s^2 - \varepsilon_2\varepsilon_1)\omega^3 d^3}{3n_s} + \frac{\omega d\varepsilon_2 + n_s}{n_s}; \quad (1b)$$

$$\frac{1-R'_f}{T_f}=\frac{\varepsilon_2{}^2\omega^4 d^4}{6}+ \\ +\frac{(\varepsilon_2-\varepsilon_2\varepsilon_1)\omega^3 d^3}{3}+\omega d\varepsilon_2+1, \quad (1c)$$

where ε_1 and ε_2 are the real and imaginary part of complex permitivity $\varepsilon = \tilde{n}^2 = \varepsilon_1 - i\varepsilon_2$, $\omega = 2\pi/\lambda$ is the wave number and n_s is the refractive index of the substrate.

In order to ease the application of the method, we perform the following algebraic transformations and substitutions in the system (1). A new variable is introduced: $V=\omega d$ and the constants are: $p_1 = (1 + R_f)/T_f$, $p_2 = (1 - R_f)/T_f$, $p_3 = (1 - R'_f)/T_f$. After this, the third equation (1c) is subtracted from the second one (1b) and the following equation is obtained:

$$C_1\varepsilon_1 + C_0 = 0, \quad (2)$$

where $C_0 = V\varepsilon_2(n_s - 1)(V^2 n_s - 3) - 3n_s(p_2 - p_3)$ and $C_1 = V^3\varepsilon_2(n_s - 1)$. Then we substitute (2) in the second equation (1b) and obtain the equation:

$$D_2\varepsilon_2{}^2 + D_1\varepsilon_2 + D_0 = 0, \quad (3)$$

where $D_2 = V^4 n_s(n_s - 1)$, $D_1 = 2V^3 n_s(n_s{}^2 - 1)$ and $D_0 = 6n_s(n_s - 1 + p_3 - n_s p_2)$.

Thus, the initial system (1) is converted into a set of three polynomials:

$$A_3\varepsilon_1{}^3 + A_2\varepsilon_1{}^2 + A_1\varepsilon_1 + A_0 = 0; \quad (4a)$$

$$C_1\varepsilon_1 + C_0 = 0; \quad (4b)$$

$$D_2\varepsilon_2{}^2 + D_1\varepsilon_2 + D_0 = 0, \quad (4c)$$

where $A_3 = -2V^4$, $A_2 = 6V^2 + 2(1 + n_s{}^2)V^4$, $A_1 = -2V^4\varepsilon_2{}^2 - 4V^3 n_s\varepsilon_2 - 6(n_s{}^2 + 1) - 2n_s{}^2V^4$ and $A_0 = \varepsilon_2{}^2(n_s{}^2V^4 + V^4 + 6V^2) + (4n_sV^3 + 12Vn_s)\varepsilon_2 + 6n_s{}^2V^2 + 6(n_s{}^2 + 1) - 12n_s p_1$.

The method of excluding the unknown quantities is applied to solve the system (4). First we exclude ε_1, using Eq. (4a) and Eq. (4b). The eliminant determinant D_{AC} of the Eq. (4a) and
Eq. (4b) is:

$$D_{AC}=\begin{vmatrix} A_3 & A_2 & A_1 & A_0 \\ C_1 & C_0 & 0 & 0 \\ 0 & C_1 & C_0 & 0 \\ 0 & 0 & C_1 & C_0 \end{vmatrix}.$$

Solving for $D_{AC} = 0$, we obtain a polynomial for ε_2:

$$E_5\varepsilon_2{}^5 + E_4\varepsilon_2{}^4 + E_3\varepsilon_2{}^3 + E_2\varepsilon_2{}^2 + E_1\varepsilon_2 + E_0 = 0, \quad (5)$$

where E_i (i = 1:5) are functions of V and n_s.

The second step is to exclude ε_2. For this purpose, Eq. (5) is used together with Eq. (4c). The eliminant determinant D_{ED} of Eq (5) and (4c) is:

$$D_{ED}=\begin{vmatrix} E_5 & E_4 & E_3 & E_2 & E_1 & E_0 & 0 \\ 0 & E_5 & E_4 & E_3 & E_2 & E_1 & E_0 \\ D_2 & D_1 & D_0 & 0 & 0 & 0 & 0 \\ 0 & D_2 & D_1 & D_0 & 0 & 0 & 0 \\ 0 & 0 & D_2 & D_1 & D_0 & 0 & 0 \\ 0 & 0 & 0 & D_2 & D_1 & D_0 & 0 \\ 0 & 0 & 0 & 0 & D_2 & D_1 & D_0 \end{vmatrix}.$$

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

$$F_5U^5 + F_4U^4 + F_3U^3 + F_2U^2 + F_1U + F_0 = 0, \quad (6)$$

where F_i (i = 1:5) are functions of n_s. Actually, this is a polynomial for d and we can find all roots of the polynomial (6) for U and all roots of d. When d is substituted in Eq. (5) or Eq. (4c), ε_2 is obtained. When ε_2 and d are substituted in Eq. (4a) or Eq. (4b), ε_1 is obtained. For verification of the accuracy of the obtained solutions, we substitute each triad (ε_1, ε_2, d) in the Eqs. (1) and the obtained results are quite close to the computer machine zero.

To illustrate the application of the described method, we make a realistic numeric simulation of spectrophotometric measurements of a nano-film, deposited on a transparent substrate, in the 450 – 800 nm spectral range. A film with values of ε_1 and ε_2 typical for the metal Au and d = 15 nm is considered. A semi-infinite substrate with ε_s very close to the values of BK7 glass is assumed. The spectral dependence of the model $\varepsilon_1(\lambda)$, $\varepsilon_2(\lambda)$ and $\varepsilon_s(\lambda)$ are presented in Fig.1.

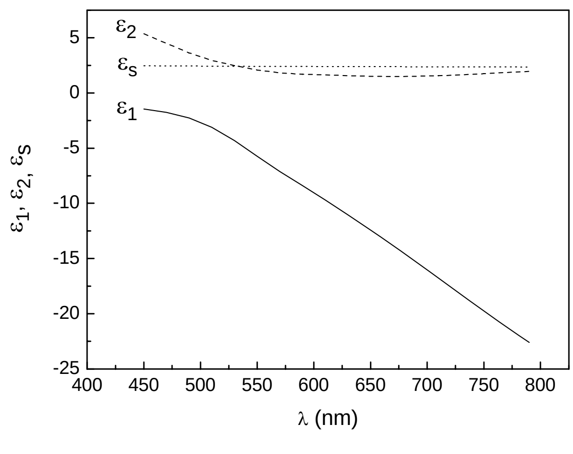

Fig. 1. Dispersion of model $\varepsilon_1(\lambda)$(—),$\varepsilon_2(\lambda)$(---) and $\varepsilon_s(\lambda)$(…).

For the needs of the inverse problem, we first evaluate for each wavelength R_f, R'_f, T_f with the help of the procedure reported in [4], and then we calculate (ε_1, ε_2, d), using the above procedure.

The polynomial (6), which is of fifth power for U, has three real positive roots for each λ. d has 6 real and 4 complex roots, but only 3 positive roots have a physical meaning. Each of these 3 roots is substituted in Eq. (5) and (4c). For the first two thicknesses $d_{1,2}$, only one value of ε_2 is simultaneously a root of Eq. (5) and (4c). For the third d_3 no coincident root of Eq. (5) and (4c) is found. This value of d is rejected. With d_1 and d_2 and the corresponding values of ε_2, we solve Eq. (4b) for ε_1.

As a measure of the accuracy of the solutions we use the relative uncertainties ($\Delta d/d$, $\Delta\varepsilon_1/\varepsilon_1$, $\Delta\varepsilon_2/\varepsilon_2$) between the model complex refractive index parameters and thickness (ε_1, ε_2, d) and the estimated values (ε_1^*, ε_2^*, d^*) [5]. The results for the spectral dependence of $\Delta\varepsilon_1/\varepsilon_1$, $\Delta\varepsilon_2/\varepsilon_2$ and $\Delta d/d$ for (ε_1, ε_2), calculated with d_1 and d_2, are presented in Fig. 2.

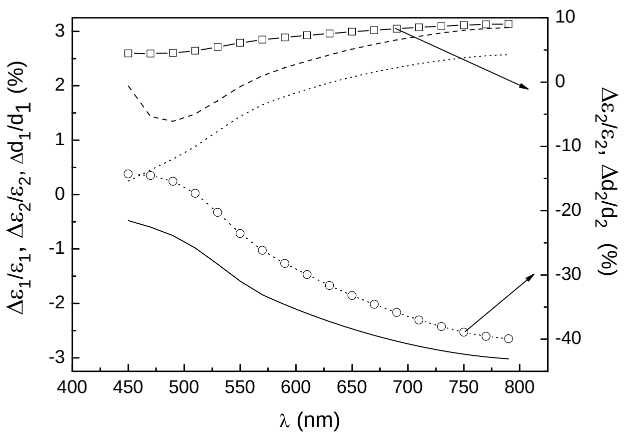

Fig.2. Dispersion of $\Delta d_1/d_1$(—), $\Delta\varepsilon_1/\varepsilon_1$(---), $\Delta\varepsilon_2/\varepsilon_2$(…) calculated with d_1 and $\Delta d_2/d_2$(–□–), $\Delta\varepsilon_2/\varepsilon_2$(–○–) calculated with d_2.

ε_1^*, ε_2^* calculated with d_1, are closer to the model values: the maximum value of $\Delta\varepsilon_1/\varepsilon_1$ is ~3%, of $\Delta\varepsilon_2/\varepsilon_2$ is ~ 2.25% and of $\Delta d_1/d_1$ is ~3%.

The values of $\Delta\varepsilon_2/\varepsilon_2$, $\Delta d_2/d_2$, calculated with d_2 are significantly bigger than those calculated with d_1. The values of ε_1^* calculated with d_2 are positive and that is why the dispersion of $\Delta\varepsilon_1/\varepsilon_1$ is not presented in Fig. 2.

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 solutions of equations (1) but because these equations are approximate.

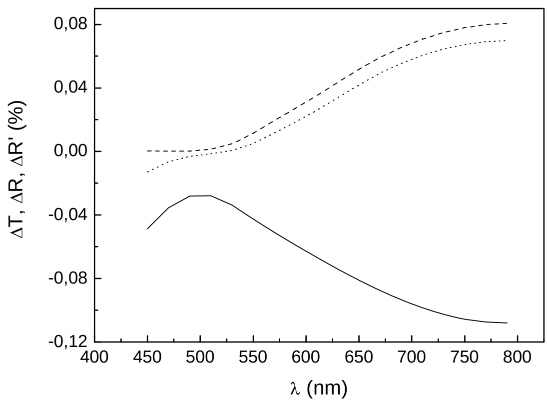

Fig. 3. Dispersion of ΔT(—), ΔR(---) and $\Delta R'$(…).

The differences $\Delta T = T_f - T_{cal}$, $\Delta R = R_f - R_{cal}$ and $\Delta R' = R'_f - R'_{cal}$ (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 the exact matrix elements) can be also used as a measure for the accuracy of the solutions. The results are presented in Fig. 3. ΔT has a maximum value of 0.12%, ΔR – 0.08% and $\Delta R'$ - 0.06%. A substantial decrease of these differences in comparison to those obtained by an expansion of the matrix elements up to third order, is observed.

As a last step in the calculations, we choose this value of $d_1(\lambda)$, at which the absolute values of ΔT, ΔR and $\Delta R'$ are minimum. In the present example, it happens at 510 nm: d_1 = 15.1 nm. Then we recalculate the ε_1 and ε_2 values with d = 15.1 nm, using Eq. (1a) and Eq. (1b), written in the form:

$$A_3\varepsilon_1^3 + A_2\varepsilon_1^2 + A_1\varepsilon_1 + A_0 = 0; \quad (7a)$$

$$B_1\varepsilon_1 + B_0 = 0; \quad (7b)$$

where $B_1 = -2V^3\varepsilon_2$, and
$B_0 = V^4 n_s \varepsilon_2^2 + 2V^3 n_s^2 \varepsilon_2 + 6d\varepsilon_2 + 6n_s(1 - p_2)$.

Their eliminant determinate D_{AB} is

$$D_{AB=}\begin{vmatrix} A_3 & A_2 & A_1 & A_0 \\ B_1 & B_0 & 0 & 0 \\ 0 & B_1 & B_0 & 0 \\ 0 & 0 & B_1 & B_0 \end{vmatrix}.$$

Solving $D_{AB} = 0$, we obtain a polynomial for ε_2:

$$I_6\varepsilon_2^6 + I_5\varepsilon_2^5 + I_4\varepsilon_2^4 + I_3\varepsilon_2^3 + I_2\varepsilon_2^2 + I_1\varepsilon_2 + I_0 = 0,$$

where I_i (i = 1:6) are functions of V and n_s. The upper equation for ε_2 has 6 roots, but only 3 of them are real and positive. These three values of ε_2 are substituted in Eq. (7b) and three values of ε_1 are calculated. Only one of the values of ε_1 is negative. This fact helps us to choose the right pair (ε_1, ε_2).

In Fig. 4, the spectral dependences of the recalculated values of $\Delta\varepsilon_1/\varepsilon_1$ and $\Delta\varepsilon_2/\varepsilon_2$ are plotted. A significant decrease of the $\Delta\varepsilon_1/\varepsilon_1$ and $\Delta\varepsilon_2/\varepsilon_2$ values in the green and red regions is seen, compared to those in Fig. 2. For instance, at 750 nm, $\Delta\varepsilon_1/\varepsilon_1$ decreases from ~2.7% to ~0.5% and $\Delta\varepsilon_2/\varepsilon_2$ - from ~2% to ~0.1%. An improvement of the values of ΔT, ΔR and $\Delta R'$ is also obtained.

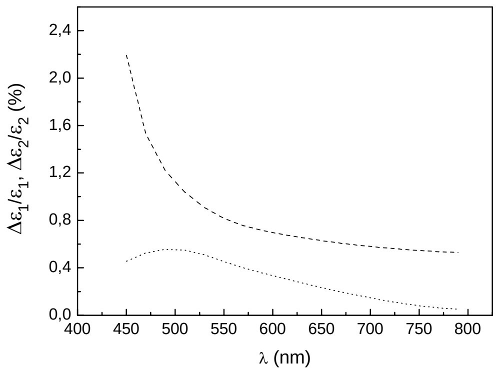

Fig. 4. Dispersion of recalculated values of $\Delta\varepsilon_1/\varepsilon_1$(---) and *$\Delta\varepsilon_2/\varepsilon_2$(…) with d_1 = 15.1 nm.*

3. Conclusion

For the first time, the system for the measurable quantities R_f, R'_f and T_f, obtained by development of the Abelès characteristic matrix elements to the 4-th power, is solved analytically. The potentialities of the approach are illustrated with a realistic example. For a thin Au film of thickness of 15 nm, the different roots of the nonlinear system are separated and those with physical meaning are easily located. A significant improvement in the determination of the Au film optical constants is obtained in the spectral range 480 – 780 nm.

Acknowledgements

This study was supported by Grant No.NPE-0404 within the frame of the National Program in Energetics.

References

[1] H. Wolter, Zeits. Physik. **105,** 269 (1937).
[2] F. Abeles, Revue D'Optique. **32,** 257 (1953).
[3] P. Gushterova, P. Sharlandjiev, B. Schmidt, M. Pham, J. Optoelectron. Adv. Mater. **7**, 305 (2005).
[4] P. Sharlandjiev, P. Gushterova, Vacuum **69**, 399 (2003).
[5] B. Hristov, P. Gushterova, P. Sharlandjiev, J. Optoelectron. Adv. Mater. **7**, 1433 (2005).

*Corresponding author: pgushterova@dir.bg