Very thin films: influence of the reflectance and transmittance uncertainties on the estimation of the films optical parameters

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Abstract. The uncertainties are investigated in estimating the optical parameters (thickness, $d$, and complex permittivity, $\varepsilon$) of very thin films arising from the methodological error and the uncertainties in the measurable quantities (transmittance, $T$, front-side reflectance, $R$, and backside reflectance, $R'$). A computer generated optical response of films with $d < \lambda/30$ ($\lambda$ being the wavelength) and dispersion with real ($\varepsilon_1$) and imaginary ($\varepsilon_2$) parts of $\varepsilon$ is used to estimate the errors in $\varepsilon$ and $d$ owing to the model approximation. A random noise with normal distribution (zero mean and variance equal to that of a high-performance spectrophotometer) is added to $T$, $R$ and $R'$ in order to study the influence of the uncertainties of $T$, $R$ and $R'$ on the calculated values of $\varepsilon_1$, $\varepsilon_2$ and $d$. It is shown that when $\varepsilon_1$, $\varepsilon_2$ and $d$ are obtained simultaneously, the influence of the two factors is considerable. The accuracy of $\varepsilon_1$, $\varepsilon_2$ and $d$ increases significantly when they are calculated in two steps, whereby the value of $d$ is first calculated followed by calculation of $\varepsilon_1$ and $\varepsilon_2$.

1. Introduction

The problem of estimating the optical parameters of very thin films, namely, the thickness, $d$, and the complex permittivity, $\varepsilon$, is challenging from a mathematical point of view and has a technological and scientific importance. To solve it, minimization techniques are often applied [1, 2, 3]. However, $\varepsilon$ and $d$ of very thin films cannot be estimated by methods that are successful for thick films. When $d/\lambda$ ($\lambda$ being the wavelength) approaches zero, the optical response of the film (transmittance, $T$, front side reflectance, $R$, and backside reflectance, $R'$) becomes less sensitive to the values of $\varepsilon$.

We have proposed [4, 5] an approach for evaluation of $\varepsilon$ and $d$ of very thin films based on limited expansion of the characteristic matrix elements up to the 4-th power with respect to $d/\lambda$. Taking the advantage of the small values of $d/\lambda$, approximate expression for $R$, $R'$ and $T$ were derived. Then $\varepsilon$ and $d$ were calculated analytically or numerically.

In the present work we analyze the uncertainties of the solutions obtained. A computer generated optical response of films with $d = 15$ nm and real, $\varepsilon_1$, and imaginary, $\varepsilon_2$, part of $\varepsilon$ very close to those of Au is used to estimate the errors in $\varepsilon$ and $d$ owing to the model approximation and due to uncertainties in $R$, $R'$ and $T$. 
2. Brief description of the determination of \( \epsilon \) and \( d \)

The approach proposed is applicable only if we have the following physical reality: a monochromatic plane wave of unit amplitude at normal incidence impinges upon a very thin optically homogeneous, isotropic film bounded by parallel surfaces. The film is supported by an optically homogeneous, isotropic semi-infinite substrate of refractive index \( n_s \). The other semi-infinite half space has a refractive index of \( n_0 = 1 \).

The optical behavior of such a film is described by its 2 x 2 characteristics matrix [6]. The nano-thickness of the film allows us to make a limited expansion of the matrix elements as a function of \( d^2/\lambda \). Retaining the terms to the fourth power we obtain expressions for \( T \), \( R \) and \( R' \). The use of the relations \( (1 + R)/T \), \( (1 - R)/T \) and \( (1 - R')/T \) leads to a set of three equations, which are simpler than those for \( T \), \( R \), \( R' \), and can be easily solved with respect to the unknown quantities \( \epsilon \) and \( d \):

\[
\frac{1 + R}{T} = \frac{(0.5\epsilon_1^2 + \epsilon_2^2 - \epsilon_2^2 \epsilon_1 + 0.5\epsilon_1^2 n_s^2 - \epsilon_1 n_s^2 + \epsilon_1^2 n_s^2 + \epsilon_2^3)\omega^4 d^4}{6n_s} + \frac{(\epsilon_2 n_s - n_s \epsilon_2 \epsilon_1)\omega^3 d^3}{3n_s} + \frac{[(1 - \epsilon_1)(n_s^2 - \epsilon_1) + \epsilon_2^2]\omega^2 d^2}{2n_s} + \frac{2\omega d \epsilon_2 n_s + (n_s^2 + 1)}{2n_s},
\]

\[
\frac{1 - R}{T} = \frac{\epsilon_2^2 \omega^4 d^4}{6} + \frac{(\epsilon_2 n_s - n_s \epsilon_2 \epsilon_1)\omega^3 d^3}{3n_s} + \frac{\omega d \epsilon_2 + n_s}{n_s},
\]

\[
\frac{1 - R'}{T} = \frac{\epsilon_2^2 \omega^4 d^4}{6} + \frac{(\epsilon_2 - n_s \epsilon_2 \epsilon_1)\omega^3 d^3}{3} + \omega d \epsilon_2 + 1,
\]

where \( \omega = 2\pi/\lambda \) is the wavenumber.

To calculate \( \epsilon_1 \), \( \epsilon_2 \) and \( d \), the Levenberg-Marquart optimization routine [4] or exact analytical approach [5] are used.

The above expressions for \( \epsilon \) and \( d \) are obtained for the case when we consider the substrate as infinite. In order to take into account the influence of the finite substrate (multiple reflections in it) and its optical performance (its small spectral absorption) we apply the corrections described in [7].

3. Estimation of the uncertainties of \( \epsilon \) and \( d \): Numeric example

To study the uncertainties of the solutions obtained we perform a realistic numeric simulation of spectrophotometric measurements of a nano-film deposited on transparent substrate in the 400 – 800 nm spectral range. A film with values of \( \epsilon_1 \) and \( \epsilon_2 \) typical for the metal Au and \( d = 15 \text{ nm} \) is considered. A semi-infinite substrate with \( \epsilon_s \) very close to the values of BK7 glass is assumed.

First, we calculate \( R \), \( R' \) and \( T \) of the film by using the exact characteristic matrix. Then we evaluate the reflectance and transmittance of the bare substrate; this is followed by obtaining the energetic coefficients of a thin film deposited onto a semi-infinite substrate: \( T_{exp} \), \( R_{exp} \), \( R'_{exp} \) [7]. In order to simulate a real measurement of the above quantities, a random noise with normal distribution (zero mean and standard deviation 0.01 of \( R \) and \( R' \) and 0.005 of \( T \)) is added. Thus ‘experimental’ front side reflectance, back side reflectance and transmittance are obtained.

To distinguish between the error due the approximation of the method and the uncertainty arising from the uncertainties in \( T \), \( R \) and \( R' \), we calculate \( \epsilon_1 \), \( \epsilon_2 \) and \( d \) in two cases: without and with noise added to \( T \), \( R \) and \( R' \). In figure 1, the spectral dependences of \( \epsilon_1 \), \( \epsilon_2 \) and \( d \), calculated without noise (black curves) and with noise (gray curves), are shown. A pronounced influence of the uncertainties of \( R \), \( R' \) and \( T \) on the calculated values of \( \epsilon_1 \), \( \epsilon_2 \) and \( d \) at \( \lambda > 500 \text{ nm} \) is observed.

As a measure of the accuracy of the solutions we use the relative uncertainties \( \Delta d/d \), \( \Delta \epsilon_1/\epsilon_1 \) and \( \Delta \epsilon_2/\epsilon_2 \) between the model complex permittivity parameters and thickness \( (\epsilon_1, \epsilon_2, d) \) and the estimated
values ($\varepsilon_1^*, \varepsilon_2^*, d^*$). The spectral dependence of $\Delta d/d$ is presented in figure 2 and these of $\Delta\varepsilon_1/\varepsilon_1$ and $\Delta\varepsilon_2/\varepsilon_2$ are given in figure 3. The black curves represent the spectral dependence of $\Delta d/d$, $\Delta\varepsilon_1/\varepsilon_1$ and $\Delta\varepsilon_2/\varepsilon_2$, calculated without noise of $R$, $R'$ and $T$. The gray curves present the spectral dependence of $\Delta d/d$ and $\Delta\varepsilon_2/\varepsilon_2$ and the red one presents the dispersion of $\Delta\varepsilon_1/\varepsilon_1$, calculated with noise added to $R$, $R'$ and $T$.

In the first case, the maximum value of $\Delta\varepsilon_1/\varepsilon_1$ is $\sim 3\%$, of $\Delta\varepsilon_2/\varepsilon_2$ is $\sim 2.25\%$ and of $\Delta d/d$ is $\sim 3\%$; whereas in the second case, the maximum value of $\Delta\varepsilon_1/\varepsilon_1$ increases to $\sim 30\%$, of $\Delta\varepsilon_2/\varepsilon_2$, to $\sim 50\%$ and of $\Delta d/d$, to $\sim 30\%$.

We estimate the value of $d$ of the film, as the mean value of $d(\lambda)$ in the spectral range, where the influence of the error due to the approximation of the method and the experimental uncertainties is minimal. In the present example, it happens between 400 nm and 500 nm and the mean value is $d = 15.1$ nm. Then we recalculate $\varepsilon_1$ and $\varepsilon_2$ values with $d = 15.1$ nm, using only equation (1a) and equation (1b). The spectral dependences of the recalculated values of $\varepsilon_1$ and $\varepsilon_2$ are shown in figure 4. The influence of the experimental uncertainties on them is appreciably reduced. Figure 5 shows plots of the dispersion of the recalculated values of $\Delta\varepsilon_1/\varepsilon_1$ and $\Delta\varepsilon_2/\varepsilon_2$. The black curves present the spectral dependence of $\Delta\varepsilon_1/\varepsilon_1$ and $\Delta\varepsilon_2/\varepsilon_2$ calculated without experimental noise added to $R$ and $T$. The gray
curves represent the spectral dependence of $\Delta \varepsilon_1/\varepsilon_1$ and $\Delta \varepsilon_2/\varepsilon_2$ calculated with experimental noise. A significant decrease is seen of the values of $\Delta \varepsilon_1/\varepsilon_1$ and $\Delta \varepsilon_2/\varepsilon_2$ in the green and red spectral regions, compared to those in figure 3. For example, at $\lambda = 750$ nm the relative error (because of the approximation of the method) of $\Delta \varepsilon_1/\varepsilon_1$ decreases from $\sim 2.7\%$ to $\sim 0.5\%$ and $\Delta \varepsilon_2/\varepsilon_2$ from $\sim 2\%$ to $\sim 0.1\%$. The influence of the uncertainties in $R$ and $T$ on $\varepsilon_2$ is negligible for $\lambda > 520$ nm. The maximal value of $\Delta \varepsilon_1/\varepsilon_1$ is $4.5\%$ at $\lambda = 450$ nm. The values of $\Delta \varepsilon_2/\varepsilon_2$ are less than $0.5\%$ in the whole spectral region.

4. Conclusion
The influence of the uncertainties of $T$, $R$ and $R'$ on the calculated values of $\varepsilon_1$, $\varepsilon_2$ and $d$ in the case of very thin films is studied. It is shown that the estimation of $\varepsilon_1$, $\varepsilon_2$ and $d$ should be performed in two steps. The first step consists in obtaining the value of $d$, while at the second step $\varepsilon_1$ and $\varepsilon_2$ are calculated. The approach proposed ensures estimation of $\varepsilon_1$, $\varepsilon_2$ and $d$ with the highest accuracy. In this case the maximal value of the relative uncertainties in $\varepsilon_1$, $\varepsilon_2$ and $d$ due to the approximation of the method and the uncertainties of the $T$, $R$ and $R'$ decreases by about a factor of ten.

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