Investigation of the uncertainties of the estimated optical constants and thickness of very thin semiconductor layers

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Abstract. We recently developed an approach to the estimation of the complex permittivity (ε) and thickness (d) of very thin layers using measurements of their transmittance, front-side reflectance and back-side reflectance. The approach is based on a limited expansion of the Abelès characteristic matrix elements and is especially designed for characterization of very thin layers. In this paper we investigate the uncertainties of the estimated real part of ε (ε₁), the imaginary part of ε (ε₂) and d of semiconductor thin layers that are due to the methodological error and the experimental uncertainties in the optical quantities measured. It is shown that the effect of the uncertainties in the measurable quantities is significantly stronger than that of the methodical error and increases considerably with the decrease of the ratio d/λ (λ being the wavelength). An efficient two-step procedure is proposed to reduce this effect. First, we supply a criterion for determination of d with the lowest uncertainty from the ensemble of estimations constructed on a wavelength by wavelength basis. This is crucial for the next step: the estimation of ε₁ and ε₂. The approach proposed ensures estimation of ε₁, ε₂ and d with the highest accuracy, limited only by the methodological error.

1. Introduction

Optically isotropic layers with a physical thickness (d) of less than λ/30 (λ being the wavelength) are key components for many applications in X-ray optics [1], microelectronics [2] and photonics [3].

The complex permittivity (ε) and d of thin layers are routinely estimated by reflectance (R) and/or transmittance (T) measurements. To evaluate d and ε, minimization techniques are most often applied [4, 5, 6]. These techniques could be divided in two broad groups: derivative-free (Simplex, direct search) methods and derivative (gradient, Levenberg–Macquardt) methods, which have several features in common: they are iterative; they need an initial guess for the model parameters; they may result in a local minimum, rather than in a global one. Using constraints on the parametric values helps but does not solve all the difficulties. Moreover, the results on very thin absorbing layers are often unreliable due to the strong correlation between ε and d [7] and the parameters obtained are susceptible to small experimental uncertainties in the optical quantities measured [8]. This is why the development of new methods for determination of ε and d, best adapted to this concrete optical problem, is still of great practical and theoretical interest.

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In several papers, we have reported a straightforward approach to the estimation of the real ($\varepsilon_1$) and imaginary ($\varepsilon_2$) parts of $\varepsilon$ and $d$ of very thin layer using measurements of the $T$, $R$ and the back-side reflectance ($R'$) of the layer. The approach is based on the limited expansion on the Abelès characteristic matrix elements and is especially designed for characterization of very thin layers [9, 10]. It possesses some important advantages: simplicity of use, no need of initial information about $\varepsilon$ and $d$, the number of the multiple solutions is significantly reduced, etc. In the present work, we investigate the uncertainties in the $\varepsilon_1$, $\varepsilon_2$ and $d$ evaluated of semiconductor thin layers. As a model layer we choose a thin film with $\varepsilon_1$ and $\varepsilon_2$ close to those of Ge because of their relatively high values in the visible and near infrared spectral ranges, and the positive values of $\varepsilon_1$ (a difficult case for evaluation of $d$ and $\varepsilon$).

2. Main equations used for the determination of $\varepsilon$ and $d$

The optical behavior of an optically homogeneous isotropic layer bounded by parallel surfaces is described by its $2 \times 2$ characteristic matrix [11]. The nano-thickness of the investigated layers allows us to make a limited expansion of the matrix elements as a function of $d$ to the 4-th power we obtain approximate 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 for the unknown quantities ($\varepsilon_1$, $\varepsilon_2$ and $d$):

$$
\frac{1 + R}{T} = \frac{(0.5\varepsilon_2^2 + \varepsilon_1^2 - \varepsilon_2^2 \varepsilon_1 + 0.5\varepsilon_2^2 \eta_2^2 - \varepsilon_2 \eta_2^2 + \varepsilon_1^2 \eta_2^2 + \varepsilon_1^3)}{6n_s} \varepsilon_1 \omega^4 d^4
$$

$$
+ \frac{(\varepsilon_2 n_s - n_\varepsilon \varepsilon_1) \omega^3 d^3}{2n_s} + \frac{[(1 - \varepsilon_1)(n_s^2 - \varepsilon_1) + \varepsilon_2 \omega^2 d^2 + 2\omega d \varepsilon_1 n_s + (n_s^2 + 1)}{2n_s},
$$

$$
\frac{1 - R}{T} = \frac{\varepsilon_2^2 \omega^4 d^4}{6} + \frac{(\varepsilon_2 n_s^2 - \varepsilon_2 \varepsilon_1) \omega^3 d^3}{3n_s} + \omega d \varepsilon_1 n_s + \frac{n_s^2}{n_s},
$$

$$
\frac{1 - R'}{T} = \frac{\varepsilon_2^2 \omega^4 d^4}{6} + \frac{(\varepsilon_2 - \varepsilon_2 \varepsilon_1) \omega^3 d^3}{3} + \omega d \varepsilon_1 + 1,
$$

where $\omega = 2 \pi \nu / \lambda$ is the wave number, $n_s$ is the substrate refractive index and $n_0$ is the refractive index of air ($n_0 = 1$).

To calculate $\varepsilon_1$, $\varepsilon_2$ and $d$, an exact analytical approach is used [12] allowing the determination of the global minima in the parameter space. We applied it to solve the above set of nonlinear equations [9].

3. Estimation of the uncertainties of $\varepsilon$ and $d$: Numerical example

The uncertainties of the solutions obtained are studied by making a realistic numerical simulation of spectrophotometric measurements of a nano-layer deposited on a transparent substrate in the 400 – 800 nm spectral range. A layer with values of $\varepsilon_1$ and $\varepsilon_2$ very close to the semiconductor Ge with $d = 15$ nm is considered. A substrate with $\varepsilon_1$ very close to the values for BK7 glass is assumed.

At the first step, $R$, $R'$ and $T$ of the layer are calculated using the exact characteristic matrix elements. Subsequently, the reflectance and transmittance of the bare substrate are evaluated, and then the energetic coefficients of a thin layer deposited onto a semi infinite substrate, $T_{exp}$, $R_{exp}$, $R'_{exp}$ are obtained [13]. A random noise with normal distribution (zero mean and standard deviation 0.01 of $R$ and $R'$ and 0.005 of $T$) is added in order to simulate a real measurement of the above quantities. Thus ‘experimental’ front-side reflectance, back-side reflectance and transmittance are generated.

Using the set of equations (1), we calculate $\varepsilon_1$, $\varepsilon_2$ and $d$ in two cases. To distinguish between the errors due to the approximation of the method and the uncertainty arising from the uncertainties in $T$,
R and R', ε₁, ε₂ and d are calculated without and with noise added to T, R and R’. Figure 1 shows the spectral dependences of ε₁, ε₂ and d, calculated without noise (black curves) and with noise (ε₂ - green curve, ε₁ - red curve and d - blue curve). A pronounced influence of the uncertainties in R, R' and T on calculated values of ε₁, ε₂ and d at λ > 620 nm is observed.

As a measure of the accuracy of the solutions we use the relative uncertainties Δd/d, Δε₁/ε₁ and Δε₂/ε₂ between the model complex permittivity parameters and thickness (ε₁, ε₂, d) and the values estimated (ε₁*, ε₂*, d*). The spectral dependences of Δd/d, Δε₁/ε₁ and Δε₂/ε₂ are presented in figure 2.

The black curves represent the spectral dependence of Δd/d, Δε₁/ε₁ and Δε₂/ε₂, calculated without noise in R, R' and T. The blue, green, and red curves present the spectral dependence of Δd/d, Δε₁/ε₁ and Δε₂/ε₂, respectively, calculated with noise added to R, R' and T. In the first case, the maximum value of Δε₁/ε₁ is ~ 8%, of Δε₂/ε₂ is ~ 11% and of Δd/d is ~ 8% at λ = 600 nm, whereas in the second case, the maximum value of Δε₁/ε₁ increases to ~ 40%, of Δε₂/ε₂ to ~ 40% and of Δd/d to ~30%.

Our calculations show that the influence of the experimental uncertainties of T, R, and R’ and the error due the approximation of the method on the calculated d, ε₁, and ε₂ can be reduced if we select one value of d(λ) and recalculate ε₁ and ε₂ values with it. The value of d for which the absolute value of Δd/d due to the method approximation is minimal is chosen as the one closest to the model one. In the present example, Δd/d has a minimal value 3.3% in the spectral region form 460 nm to 500 nm, where d is 14.5 nm. With this value, which fixes the physical thickness, we recalculate ε₁ and ε₂ using (1a) and (1b) only. The spectral dependences of the recalculated values of ε₁ and ε₂ are shown in figure 3.
It is seen that the influence of the experimental uncertainties on them is appreciably reduced. In figure 4, the dispersion of the recalculated values $\Delta \varepsilon_1/\varepsilon_1$ and $\Delta \varepsilon_2/\varepsilon_2$ are plotted. 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 colored curves represent the spectral dependence of $\Delta \varepsilon_1/\varepsilon_1$ (green) and $\Delta \varepsilon_2/\varepsilon_2$ (red), calculated with experimental noise. The significant decrease of the values of $\Delta \varepsilon_1/\varepsilon_1$ and $\Delta \varepsilon_2/\varepsilon_2$ due to the uncertainties in the measured $R$, $R'$ and $T$, compared to those in figure 2 is obvious. The methodological error is also slightly reduced. For example, at $\lambda = 600$ nm, the relative error (due to the approximation of the method) of $\Delta \varepsilon_1/\varepsilon_1$ decreases from $\sim$ 8% to $\sim$ 3.8%, and of $\Delta \varepsilon_2/\varepsilon_2$ – from $\sim$ 11.5% to $\sim$ 7.6%.

4. Conclusion

A straightforward approach for the determination of $\varepsilon_1$, $\varepsilon_2$ and $d$ of very thin layers is applied to determine $\varepsilon_1$, $\varepsilon_2$ and $d$ of semiconductor layers. The influence of the methodological error and the uncertainties in $T$, $R$ and $R'$ on the calculated values of $\varepsilon_1$, $\varepsilon_2$ and $d$ are analyzed. The calculations performed show that the effect of the uncertainties in the measurable quantities is significantly higher than that of the methodological error and increases considerably with the decrease of the $d/\lambda$ ratio. The accuracy of the solution increases by about a factor of ten if the value of $d$ is estimated first and then the values of $\varepsilon_1$ and $\varepsilon_2$ are recalculated in the whole spectral region under investigation.

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