Improvement of the measurement accuracy of the spectral method for evaluation parameters of the optically transparent thin films

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Abstract. In this paper the ways of increasing the measurement accuracy of the parameters of the optically transparent thin films using the spectral method are described. The proposed plotting of the envelope curves for the minima and maxima of the interference oscillations in a film and additional filtering of the spectral data, allows improving the accuracy of determining the extrema position and, consequently, increasing the accuracy of the thickness measurements of the deposited coatings.

Thin films and coatings are widely used in industry and household appliances and devices. Control of the parameters of such coatings should be conducted using the non-destructive methods, the most suitable of which is the spectral method [1, 2].

The process of radiation propagation in thin films and related effects are well studied and described. Registered with the spectrometer passed through the film or reflected from the film radiation has oscillations, characteristics of which depend on the thickness and refractive index of the film and the substrate material on which the film is applied [3, 4]. Having information about the film and the substrate materials, and knowing, therefore, their refractive indices it is possible to calculate the film thickness according to the position of the neighboring extrema. However, this method has a significant drawback which is the difficulty of determining the exact position of the extrema of the oscillations in the real noisy spectrum, especially when the control of the film parameters is conducted in the conditions of industrial production.

Industrial manufacture of products with film coating means a rather high reproducibility of their parameters, and, consequently, the known refractive indices of the used materials. This fact allows increasing the calculation accuracy of the measured film parameters through the use of the theoretical calculations of the reflectance or transmittance spectrum shape. For measurements of film parameters in real time can be used a compact spectrometer with fiber-optic input of the radiation and an optical scheme of the experimental installation, allowing to carry out measurements both on reflection and transmission, depending on the type of a substrate [5, 6]. Parameters of the spectrometer must allow measurement of the transmittance or reflectance in a wide wavelength range; meanwhile there are no special requirements on the spectral resolution of the device, since the extrema of the recorded spectrum are obviously separated from each other by more than a few nanometers.

During the passage of monochromatic radiation through a thin film occurs interference, which in a broad spectral range looks like oscillatory modulation of the spectral signal. The theoretical form of
the spectrum for the film of a given thickness with known refractive index at normal incidence of radiation can be modeled based on expressions for the transmission \( T \) and reflection \( R \) coefficients [7]:

\[
T = \frac{n_1}{n_0} \left( \frac{t_1^2 t_2^2}{1 + 2 r_1 r_2 \cos 2 \delta_1 + r_1^2 r_2^2} \right); \tag{1}
\]

\[
R = \left( \frac{r_1^2 + 2 r_1 r_2 \cos 2 \delta_1 + r_2^2}{1 + 2 r_1 r_2 \cos 2 \delta_1 + r_1^2 r_2^2} \right), \tag{2}
\]

where

\[
\delta_1 = \left( nd \frac{2\pi}{\lambda} \right); t_1 = \left( \frac{2n_0}{n_0 + n} \right); t_2 = \left( \frac{2n}{n + n_z} \right); r_1 = \left( \frac{n_0 - n}{n_0 + n} \right); r_2 = \left( \frac{n - n_z}{n + n_z} \right), \tag{3}
\]

here

\[
d = \text{film thickness}; \quad n_0 = \text{refractive index of the air}; \quad n = \text{refractive index of the film}; \quad n_z = \text{refractive index of the substrate}, \quad \text{provided that the values of refractive indices } n > n_0; \quad n > n_z.
\]

In the general case, the refractive indices of the film and substrate materials have dispersion dependent on the wavelength of the radiation. Therefore, in expressions (3) it is necessary to use not constants, but the values calculated for each wavelength.

For calculation of the film parameters the spectrometer management program ASpect2010 [5] has been modified – was added calculation of the theoretical shape of spectrum using expressions (1)–(3) for a given film thickness with a known refractive index. The program has a database of most common materials used for film deposition, taking into account the dispersion of the refractive index (figure 1).

**Figure 1.** Choice of a film material from the Aspect2010 database.

**Figure 2.** Simulation of the reflectance of SiO\(_2\) film with a thickness of 1 µm.

Step between the points and their number for each material is different, and for correct calculations of \( T \) and \( R \) for each material approximation is performed for the specified points to obtain the intermediate values. For approximation cubic splines are used because they provide a continuous first and second derivatives, which guarantees the continuity of the plotting of the spectral distribution of \( T \) and \( R \). Comparison of simulation results of the reflection coefficient of the SiO\(_2\) films in the program ASpect2010 (figure 2), and using the widely known Filmetrics site [8] gives almost identical results, confirming the correctness of the implemented calculation procedures.

The maxima and minima of \( T \) and \( R \) are recorded under certain conditions. For example, for \( T_{\text{min}} \) can be written

\[
d = \frac{\lambda}{4} (2m + 1), \tag{4}
\]

where \( m = 0, 1, 2, ... \) – order of the minimum.

For \( T_{\text{max}} \), the expression would be as follows

\[
d = \frac{\lambda}{4} (2m + 2). \tag{5}
\]
For calculation of the film thickness it is necessary to transform expressions (1), (2) for the values of the extrema of transmittance and reflectance [9]:

\[
T_{\text{min}} = \left( \frac{4n_0n_1n_2}{(n_0n_2 + n_1^2)} \right); \\
T_{\text{max}} = \left( \frac{4n_0n_2}{(n_0 + n_2^2)} \right);
\]

\[(6)\]

\[
R_{\text{min}} = \left( \frac{n_0 - n_1}{n_0 + n_2} \right)^2; \\
R_{\text{max}} = \left( \frac{n_0^2 - n_1n_2}{n_0^2 + n_1n_2} \right)^2.
\]

\[(7)\]

From (6), (7) can be obtained the expressions for calculation of the refractive index of a film:

\[
n = \sqrt{n_1n_2}\left( \frac{1 + \sqrt{1 - T_{\text{min}}}}{T_{\text{min}}} \right); \\
n = \sqrt{n_0n_2} \left( 1 + \sqrt{\frac{1 + R_{\text{max}}}{1 - R_{\text{max}}} - 1} \right).
\]

\[(8)\]

Thus, it is necessary to determine the position of extrema in the spectrum of the reflectance or transmittance of a film. To obtain the data on the actual index of refraction in the whole spectral range in the program are plotted the envelope curves for maxima and minima of the spectra (figure 3).

![Figure 3. Reflection spectrum of the Ta_2O_5 film with plotted envelope curves for maxima and minima.](image)

When building envelope curves it must be considered that the number of extrema depends on the film thickness and can be small. To plot the envelope curves across the entire spectral range to the found extrema are added the additional counts on the edges of the band. If there is only one extremum the envelope curve is given as a horizontal line, in the presence of two extrema the envelope curve is defined as the line passing through them, if there are three or more extrema the end points are specified using linear interpolation on the nearest to each edge pair of extrema. These values are approximated by a third degree polynomial and are displayed on the screen together with the original spectra, which allows to visually verify the correctness of building envelope curves.

Then using the constructed envelope curves with the help of (8), depending on the type of spectrum is plotted the dependence of the refractive index of a film from the wavelength. Taking into account (4), (5) the expression for calculation of the film thickness takes the form

\[
d = \frac{\lambda_{\text{e}1}\lambda_{\text{e}2}}{2(\lambda_{\text{e}1}n_{\text{e}2} - \lambda_{\text{e}2}n_{\text{e}1})},
\]

where \(\lambda_{\text{e}1}\) – wavelength of the first extremum; \(\lambda_{\text{e}2}\) – wavelength of the second extremum; \(n_{\text{e}1}\) – refractive index of the film in the first extremum; \(n_{\text{e}2}\) – refractive index of the film in the second extremum.
The “classic” method of finding the maximum or minimum in the spectra of reflection or transmission leads to the significant errors due to the presence of noise on the fairly flat extrema. For example, two of the reflection spectrum of the same film of SiO$_2$ on a glass substrate (figure 4(a)) in search for one of the peaks gave results 723.6 and 730.7 nm. Such a large variation in the measurement of the extremum position leads to the serious errors of measurement of the film thickness.

Figure 4. Measuring of the position of one of the peaks on SiO$_2$ film spectrum with different limits of integration: (a) – before filtering of the signal; (b) – after filtering of the signal.

For increasing the measurement accuracy by reducing the effect of noise can be used the calculation of the center of gravity for the peak, which is carried out according to the expression

$$\lambda_c = \frac{\sum \lambda_i I_i}{\sum I_i}$$

(9)

where $i$ – element number from the array of spectral data from the left to the right border of the peak (are taken into account only the elements of the array greater than the given level); $\lambda_i$ – wavelength of the $i$-th element of the spectrum; $I_i$ – amplitude of the $i$-th element of the spectrum.

The values of the wavelength for the center of gravity of the peak $\lambda_c$ (figure 4(b)) are 727.9 and 728.2 nm, so the dispersion is about 20 times less than for conventional extremum seeking. However, must be kept in mind the fact that the extremes are not symmetrical. This leads to the changes in results when moving the boundaries of numerical integration according to (9).

For minimization of the additional errors associated with the choice of boundaries their position should be always selected on the same level. When narrowing the boundaries of the integration distance between the maximum value and center of gravity of the extremum $\lambda_c$ value is reduced to 0.5 nm from the initial distance of 5 nm for the widest possible area of integration. For the experimentally found level of 20 % of the difference between neighboring extrema the received location values of the extrema with the minimum error correspond to the theoretically calculated values. While calculating film thickness as a first approximation can be taken the value of the film thickness, obtained for any two neighboring extrema. To improve the measurement accuracy the filtering of a signal is conducted, however the thickness value will be clearly erroneous, because in the presence of noise it is impossible to accurately determine the position of extremum. Improvement of the measurement accuracy for the calculation of film thickness is achieved at the following stage. The cumulative absolute value of the difference between the samples of the theoretical and real spectra is calculated:

$$D = \sum_{\lambda=180}^{1080} I_{ex} - \sum_{\lambda=180}^{1080} I_{th},$$

where $I_{ex}$ – count values of the experimental spectrum; $I_{th}$ – count values of the theoretical spectrum. The wavelength range 180...1080 nm can be reduced depending on the actual parameters of the device.

The presence of noise in the real spectrum means that the calculated integrated difference with the theoretical spectrum will never be zero, but it can be minimized bringing the shape of the theoretical
spectrum closer to the shape of a real spectrum. For this should be built several theoretical spectra for thicknesses that are close to the initial calculated thickness, in a range, obviously exceeding the measurement error. For each of the plotted theoretical spectra it is necessary to calculate the integral difference with the real spectrum and to compare them with each other. As a result a spectrum with a minimum integral difference between the real and theoretical spectrum will give the most accurate result of the film thickness (figure 5).

Figure 5. Theoretical spectrum with the minimum difference from the real spectrum, calculation of the film thickness.

Thus, the conducted research and developed methodology allowed to minimize the influence of the real spectrum noise on the measurement results of the film parameters, greatly increasing the accuracy of calculations. However, it should be noted that in the absence of reliable information on the film material and the refractive index shape of the theoretical spectrum will differ significantly from the real one. This circumstance does not allow obtaining the exact value of the film thickness by the proposed method, since the refractive index of the films influences not only the period of the oscillations, but also their amplitude.

Measurement of the thin films parameters by proposed spectral method includes the following steps:

- acquisition of the reflectance or transmittance spectrum;
- normalization of the spectrum on a radiation source (calculation of the reflectance or transmittance coefficient);
- filtering of the obtained reflectance or transmittance coefficient;
- calculation of the refractive index of the film and substrate;
- selection of the extrema for pre-calculating of the film thickness;
- generation of a model of reflectance or transmittance based on the conducted measurements;
- refinement of the film thickness value for obtaining the best fit of the model to the real reflectance or transmittance coefficient.

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