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Calculation of the optical constants of a thin layer upon a transparent substrate from the reflection spectrum

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Abstract. A method has been proposed for calculation of the optical constants of a thin layer upon a transparent substrate only from the reflection spectrum into the region of transmission of the layer. The envelopes of the maxima of the spectrum R_M and of the minima R_m are used, taking into account the finite dimensions of the substrate. The algorithm of the calculations is similar to the one proposed by Swanepoel [9] using the transmission spectrum. The method makes it possible to calculate the thickness of the layer and the refractive index n with an error of 0.2%. The correct description of the dispersive relationship of n is obtained for an amorphous layer $Ge_{19}As_{21}S_{60}$.

1. Introduction

The main method for calculating the optical constants of thin layers is based on scanning the transmission and reflection spectra of a thin layer from the material under investigation laid down upon a thick transparent substrate. The rapid oscillation of the spectra with changing wavelength, however, leads to non-singular solutions of the system containing two equations being obtained—one for transmission T and one for reflection R with respect to the refractive and absorption indices n and k of the layer [1–3].

There are two ways of eliminating the non-singularity of the solution without using complex iteration procedures that always lead to the use of monotonic functions of λ for calculating the optical constants of the layer. The first way makes use of different combinations of the transmission and reflection spectra [4–6]. The second way uses their envelopes, which are monotonic functions of λ [7], instead of the spectra themselves.

Methods for calculating n and k from the envelopes of the transmission spectrum alone are presented by Manifacier [8] and Swanepoel [9]. In the first case the substrate is considered to be semi-infinite, while in the second its final dimensions are taken into account.

Kushev and Jeleva [10] have proposed a method of calculation using the envelopes of the reflection spectrum. They obtained simple analytical expressions for n and k, but the substrate is again considered to be semi-infinite.

The present article proposes a method for calculating n and k of the layer from the envelopes of the reflection spectrum, taking into account the finite dimensions of the substrate.

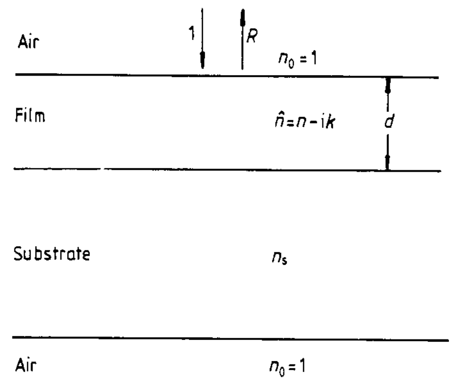

Figure 1. Diagram showing the reflection of light incident normal to the surface of a thin homogeneous layer of thickness d, refractive index n and absorption index k upon a thick, finite and transparent substrate of refractive index n_s.

2. Theory

A diagram of the case under consideration—light incident normally on the surface of a thin, homogeneous layer with a constant thickness d—is shown in figure 1. The reflection of this system when the layer transmits the light ($T > 0$), and the refractive index of the layer, n, is bigger than that of the substrate n_s, is calculated by the well known formula [11]

$$R = \rho_{13}^2 + \frac{n_s^2 \tau_{13}^4 \rho_{34}}{1 - \rho_{31}^2 \rho_{34}^2} \quad (1)$$

where τ_{ij} and ρ_{ij} are the amplitude coefficients of transmission and reflection between the media i and j, the indices 1 and 4 refer to the air, 2 to the layer and 3 to the substrate.

The above-mentioned amplitude coefficients of transmission and reflection between non-adjacent media are calculated using the expressions

$$\tau_{13}^2 = \frac{\tau_{12}^2 \tau_{23}^2}{x^{-1} + \rho_{12}^2 \rho_{23}^2 x - 2\rho_{12}\rho_{23} \cos \Delta_2} \quad (2a)$$

$$\rho_{13}^2 = \frac{\rho_{12}^2 x^{-1} + \rho_{23}^2 x - 2\rho_{12}\rho_{23} \cos \Delta_1}{x^{-1} + \rho_{12}^2 \rho_{23}^2 x - 2\rho_{12}\rho_{23} \cos \Delta_2} \quad (2b)$$

$$\rho_{31}^2 = \frac{\rho_{12}^2 x + \rho_{23}^2 x^{-1} - 2\rho_{12}\rho_{23} \cos \Delta_1'}{x^{-1} + \rho_{12}^2 \rho_{23}^2 x - 2\rho_{12}\rho_{23} \cos \Delta_2} \quad (2c)$$

where $\Delta_1 = -\Delta_{12} + \Delta_{23} + 2\delta$; $\Delta_1' = \Delta_{12} - \Delta_{23} + 2\delta$; $\Delta_2 = \Delta_{12} + \Delta_{23} + 2\delta$; $\alpha = 4\pi k/\lambda$; $x = \exp(-\alpha d)$; $\delta = 2\pi nd/\lambda$ and Δ_{ii+1} are the phase differences which appear when the light passes through the boundary between the adjacent media.

The amplitude coefficients of transmission and reflection and the phase differences which arise at the boundaries between the adjacent media are determined from

$$\tau_{12} = 2/[(1+n)^2 + k^2]^{1/2}$$

$$\tau_{23} = 2\{(n^2 + k^2)/[(n+n_s)^2 + k^2]\}^{1/2}$$

$$\tau_{34} = 2n_s/(n_s + 1)$$

$$\rho_{12} = \{[(1-n)^2 + k^2]/[(1+n)^2 + k^2]\}^{1/2}$$

$$\rho_{23} = \{[(n-n_s)^2 + k^2]/[(n+n_s^2) + k^2]\}^{1/2}$$

$$\rho_{34} = (n_s - 1)/(n_s + 1)$$

$$\Delta_{12} = \tan^{-1}[2k/(n^2 + k^2 - 1)]$$

$$\Delta_{23} = \tan^{-1}[2n_s k/(n^2 + k^2 - n_s^2)].$$

On the basis of the expressions (2), equation (1) can be represented as a function of the refractive and absorption indices of the layer:

$$R = \frac{A' - (B_1' \cos 2\delta - B_2' \sin 2\delta)x + C'x^2}{A'' - (B_1'' \cos 2\delta - B_2'' \sin 2\delta)x + C''x^2} + \frac{A'''x^2}{[A'' - (B_1'' \cos 2\delta - B_2'' \sin 2\delta)x + C''x^2]} \times \frac{1}{[D'' - (E_1'' \cos 2\delta - E_2'' \sin 2\delta)x + F''x^2]} \quad (3)$$

where

$$A' = [(n-1)^2 + k^2][(n+n_s)^2 + k^2]$$

$$B_1' = 2[(n^2+k^2-1)(n^2+k^2-n_s^2) + 4k^2 n_s]$$

$$B_2' = 4k[n_s(n^2+k^2-1) - (n^2+k^2-n_s^2)]$$

$$C' = [(n+1)^2 + k^2][(n-n_s)^2 + k^2]$$

$$A'' = [(n+1)^2 + k^2][(n+n_s)^2 + k^2]$$

$$B_1'' = 2[(n^2+k^2-1)(n^2+k^2-n_s^2) - 4k^2 n_s]$$

$$B_2'' = 4k[n_s(n^2+k^2-1) + (n^2+k^2-n_s^2)]$$

$$C'' = [(n-1)^2 + k^2][(n-n_s)^2 + k^2]$$

$$A''' = 64 n_s (n_s - 1)^2 (n^2 + k^2)^2$$

$$D'' = [(n+1)^2 + k^2][(n+1)(n+n_s^2) + k^2]$$

$$E_1'' = 2[(n^2+k^2-1)(n^2+k^2-n_s^2) - 2k^2(n_s^2+1)]$$

$$E_2'' = 2k[(n^2+k^2-n_s^2) + (n_s^2+1)(n^2+k^2-1)]$$

$$F'' = [(n-1)^2 + k^2][(n-1)(n-n_s^2) + k^2].$$

Since in the spectral region in question $n > n_s \gg k$, k can be approximated by 0 in equations (1)–(3) and the reflection of the system now becomes a function of n and x only. Then the envelopes of the reflection spectrum maxima R_M and minima R_m are represented by (1), when $2\delta = \pi$ and $2\delta = 0$ are respectively substituted into (2).

Equation (3) then has the following form:

$$R_M, R_m = \frac{A' \pm B_1'x + C'x^2}{A'' \pm B_1''x + C''x^2} + \frac{A'''x^2}{(A'' \pm B_1''x + C''x^2)(D'' \pm E_1''x + F''x^2)} \quad (4)$$

where + in the $\pm$ refers to R_M and $-$ to R_m, or

$$R_M, R_m = \frac{(ad \pm bcx)^2}{(bd \pm acx)^2} + \frac{gx^2}{(bd \pm acx)^2(b^3 f \pm 2abcdx + a^3 ex^2)} \quad (5)$$

where $a = n - 1$, $b = n + 1$, $c = n - n_s$, $d = n + n_s$, $e = n - n_s^2$, $f = n + n_s^2$, $g = 64 n_s (n_s - 1)^2 n^4$.

In the transparent region of the layer $x = 1$ and (5) referring to the minimum leads to the well known equation

$$R_m = \frac{(n_s - 1)^2}{n_s^2 + 1}$$

which can serve for determination of n_s:

$$n_s = \frac{1 + [R_m(2 - R_m)]^{1/2}}{1 - R_m}. \quad (6)$$

In this case the problem of calculating n and k for the layer for all λ reduces to solving a system of two equations for R_M and R_m for the unknowns n and k according to (5).

On the basis of the approximation $k = 0$, the order number of a given extremum from the spectrum m is

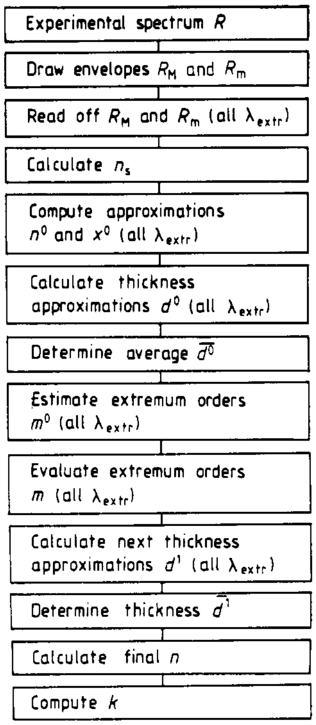

Figure 2. Block diagram of the algorithm for calculating $n(\lambda)$ and $k(\lambda)$ of a thin layer from the reflection spectrum. The symbols are defined in § 3, where a full explanation of the algorithm is given.

determined from:

$$4nd = m\lambda_{extr} \tag{7}$$

and an odd order number m corresponds to λ_{extr}, for which the reflection spectrum has a maximum λ_{max}, and an even order number m^- to λ_{min}.

This being the case, the thickness of the layer can be determined by the known wavelengths λ_1 and λ_2 and the respective refractive indices of the layer n_1 and n_2 for two adjacent extrema

$$d = \frac{\lambda_1 \lambda_2}{4(\lambda_1 n_2 - \lambda_2 n_1)}. \tag{8}$$

3. Algorithm of the calculation

A block diagram of the algorithm for calculating the spectral dependences $n(\lambda)$ and $k(\lambda)$ of a thin layer using only the reflection spectrum is given in figure 2. The essence of the algorithm is in the exact determination of the order numbers m of the extrema and the thickness of the layer, based on the approximated values of the refractive index n^0 and the thickness of the layer d^0, and hence the values of n and k. The calculations are performed in the same sequence as in the method proposed by Swanepoel [9], where the transmission spectrum is used.

It is necessary to give some explanation about the way the algorithm is used. The refractive index of the substrate n_s is calculated from the envelope R_m in the spectral region of transparency of that layer from (6). The Newton–Raphson iteration can be used for solving the system of two equations (5) for R_M and R_m referring to the two unknown approximated values n^0 and x^0. In order to achieve faster convergence, the calculations are best begun for λ_{extr} from the transparent region, where for an initial approximation of n^0 a presumed value is used, and for $x^0 \rightarrow x^0 = 1$. Since λ_{extr} decreases for the initial approximations one has to use n^0 and x^0 calculated for the previous extremum.

The approximated values for the thickness of the layer d^0 for every λ_{extr} are calculated from the data for n^0 of the previous extremum and neighbouring one (equation (8)), and $\overline{d^0}$ is the average value of d^0 for all λ_{extr}. The order of a given extremum m^0 can be estimated from (7) using $\overline{d^0}$ and the corresponding n^0. The orders m of the neighbouring extrema are in fact consecutive integers, even for the minima and odd for the maxima of the reflection. The calculation of the next thickness approximation d^1 is done via (7) where n^0 and m are used, while $\overline{d^1}$ is the average value of the thickness of the layer for all λ_{extr}. $\overline{d^1}$ is the calculated thickness of the layer.

Substituting $\overline{d^1}$ and m in equation (7) the final value of n for each extremum is obtained. The value of k for each n is found by solving equation (3), written for one of the envelopes R_M and R_m at a given n, for k_1 by the Newton iteration.

4. Accuracy of the method

To appreciate the accuracy of the method for calculating the indices of refraction and absorption of a thin layer upon a finite transparent substrate, the reflection spectrum at given parameters of the system drawn in figure 3 is solved from equation (3). The

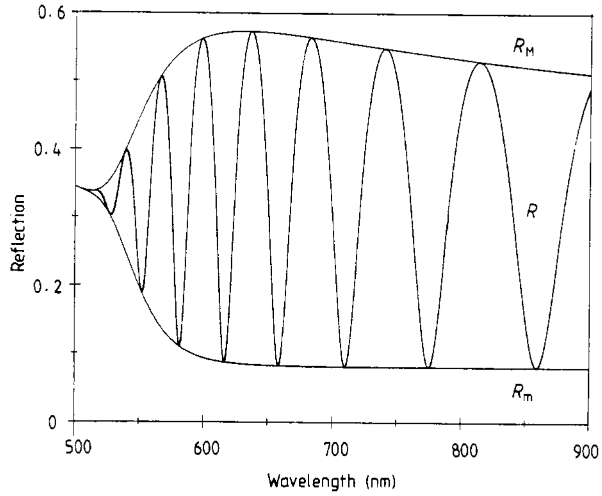

Figure 3. Reflection spectrum of a thin layer with a thickness $d = 1000$ nm laid upon a substrate with a refractive index $n_s = 1.51$ (constant). The spectral behaviour of the refractive and absorption indices of the layer are $n_{tr} = 3 \times 10^5/\lambda^2 + 2.6$ and $k_{tr} = \lambda/4\pi \times 10^{[(1.5 \times 10^6/\lambda^2)-8]}$ (λ in nm).

Table 1. Calculation of the spectral behaviour of the refractive index $n(\lambda)$ and the absorption index $k(\lambda)$ of a layer with $n_{tr} = 3 \times 10^5/\lambda^2 + 2.6$ and $k_{tr} = \lambda/4\pi \times 10^{[(1.5\times10^6/\lambda^2)-8]}$ (λ in nm) from the reflection spectrum shown in figure 3. The other symbols are defined in § 3 and § 4.

λ (nm)	n_{tr}	k_{tr}	n^0	x^0	d^0 (nm)	m^0	m	d^1 (nm)	n	k (using envelope R_M)	k (using envelope R_m)
859	3.007	7.373×10^{-5}	2.995	1.008	1008	13.9	14	1004	3.007	1.629×10^{-4}	-5.510×10^{-4}
814	3.053	1.189×10^{-4}	3.040	1.008	874	14.9	15	1004	3.053	1.860×10^{-4}	-4.879×10^{-4}
775	3.099	1.939×10^{-4}	3.116	0.985	1012	16.0	16	995	3.100	1.962×10^{-4}	9.329×10^{-4}
740	3.148	3.220×10^{-4}	3.158	0.986	1032	17.0	17	996	3.145	2.727×10^{-4}	8.473×10^{-4}
710	3.195	5.341×10^{-4}	3.202	0.987	1089	17.9	18	998	3.195	4.952×10^{-4}	7.716×10^{-4}
683	3.243	8.927×10^{-4}	3.237	0.987	988	18.9	19	1002	3.244	9.894×10^{-4}	7.073×10^{-4}
659	3.292	1.505×10^{-3}	3.290	0.972	936	19.9	20	1002	3.295	1.654×10^{-3}	1.483×10^{-3}
636	3.341	2.573×10^{-3}	3.345	0.947	1068	20.9	21	998	3.339	2.531×10^{-3}	2.745×10^{-3}
616	3.390	4.363×10^{-3}	3.384	0.917	965	21.9	22	1001	3.388	4.385×10^{-3}	4.237×10^{-3}
598	3.439	7.449×10^{-3}	3.440	0.854	957	22.9	23	1000	3.439	7.487×10^{-3}	7.535×10^{-3}
581	3.488	0.0127	3.494	0.758	1054	23.9	24	998	3.486	0.0126	0.0129
566	3.536	0.0217	3.538	0.617	1011	24.9	25	1000	3.538	0.0217	0.0217
552	3.586	0.0372	3.587	0.429	1025	25.9	26	1000	3.588	0.0373	0.0371
539	3.634	0.0635	3.634	0.228	984	26.8	27	1001	3.638	0.0638	0.0632
526	3.683	0.1085	3.680	0.075	920	27.8	28	1001	3.682	0.1098	0.1077
515	3.731	0.1854	3.743	0.011	—	—	29	998	3.734	0.1869	0.1854

$n_s = 1.51$; $\overline{d^0} = 995$ nm; $\overline{d^1} = 1000$ nm

results from the calculation of n and k from the envelopes R_M and R_m of this spectrum in accordance with the algorithm described in the previous paragraph are shown in table 1. The last two columns of the table contain the calculated values for k at a known n, from the envelopes R_M and R_m respectively. The comparison of the data for k and k_{tr} shows that more exact values for k are obtained by using the envelopes R_M than R_m, and the difference is mainly in the region of weak absorption where $x \simeq 1$. This can be explained by the fact that R_m is independent of n and k in this region—see equation (6). The accuracy which is achieved with the calculation of n in the whole spectrum ($(n - n_{tr})/n_{tr} \leqslant 0.2\%$) is better than that for calculation of k($(k - k_{tr})/k_{tr} \leqslant 1.5\%$) in the region of medium and strong absorption ($x < 0.9$) which is related to the stronger sensitivity of R towards changes in n than in k. The considerable inaccuracy which is present during calculation of k in the region of weak absorption is due to the smaller value of k in this region. That is why R_M is nearly insensitive to changes in k.

Another important fact is that in the method proposed by Swanepoel [9] for calculating n and k from the transmission spectrum, the short wave extrema give

Table 2. Calculated values for the refractive and absorption indices of a thin amorphous layer $Ge_{19}As_{21}S_{60}$, using an experimentally obtained reflection spectrum (§ 5). Symbols as defined in § 3.

λ (nm)	R_M	R_m	n^0	x^0	d^0 (nm)	m^0	m	d^1 (nm)	n	k (using envelope R_M)
937	0.367	0.067	2.384	1.009	1065	12.8	13	1277	2.388	$\simeq 0$
875	0.371	0.067	2.398	1.008	1274	13.8	14	1277	2.402	$\simeq 0$
822	0.376	0.067	2.414	1.008	1274	14.8	15	1277	2.418	$\simeq 0$
775	0.380	0.067	2.428	1.007	1238	15.7	16	1277	2.431	$\simeq 0$
734	0.384	0.067	2.442	1.007	1276	16.7	17	1277	2.447	1.366×10^{-5}
698	0.389	0.067	2.459	1.006	1324	17.7	18	1277	2.464	2.620×10^{-5}
666	0.393	0.067	2.472	1.006	1309	18.6	19	1280	2.481	2.080×10^{-5}
637	0.397	0.067	2.486	1.006	1057	19.6	20	1281	2.498	3.647×10^{-4}
611	0.400	0.068	2.529	0.967	1433	20.8	21	1268	2.516	7.092×10^{-4}
587	0.401	0.068	2.532	0.967	1257	21.7	22	1275	2.532	1.242×10^{-3}
566	0.401	0.069	2.554	0.941	1283	22.7	23	1274	2.553	2.101×10^{-3}
546	0.396	0.071	2.573	0.900	1271	23.7	24	1273	2.569	3.455×10^{-3}
528	0.386	0.075	2.592	0.837	1299	24.7	25	1273	2.588	5.674×10^{-3}
512	0.368	0.083	2.612	0.744	1250	25.6	26	1274	2.610	9.388×10^{-3}
494	0.329	0.102	2.619	0.576	1295	26.6	27	1273	2.615	0.0169
475	0.267	0.140	2.610	0.317	1217	27.6	28	1274	2.608	0.0340
457	0.214	0.183	2.605	0.077	1233	28.6	29	1272	2.599	0.0722
441	0.199	0.197	2.603	0.005	—	29.7	30	1271	2.594	0.1481

$n_s = 1.458$; $\overline{d^0} = 1256$ nm; $\overline{d^1} = 1275$ nm a false value for the thickness of the layer and is not taken into account in the calculations, but in our method it is quite different. As a consequence, when one deals with the reflection spectrum a greater number of extrema are used, which leads to reduced errors when determining d and the order numbers of the corresponding extrema.

5. Experiment

The method proposed above is used for calculating the thickness of a sample of a vacuum-evaporated amorphous layer of $Ge_{19}As_{21}S_{60}$ on a quartz substrate. The reflection spectrum is scanned on a Perkin–Elmer 330 spectrophotometer. The results from the calculations are shown in table 2. An important fact is the presence of a maximum in the function $n(\lambda)$ while k increases with the decrease of λ to $T = 0$. A similar behaviour of the optical constants was observed for the amorphous chalcogenides with composition Ge_2S_3 [12] and $Ge_xAs_2S_3$ ($x = 0$–4) [13] and can be explained by the low coordination number and the presence of lone-pair orbitals, which lead to blurring of the edges of the zone diagrams.

Calculations of $n(\lambda)$ of a thin layer with composition $Ge_{19}As_{21}S_{60}$ from the envelopes of the *transmission* spectrum according to [9] are made in [14] for $\lambda > 550$ nm. The difference between these values and the ones given in table 2 for n does not exceed 0.6%, which shows a very good agreement between the calculated values for n from the envelopes of the transmission and reflection spectra.

6. Conclusion

A method for calculating $n(\lambda)$ and $k(\lambda)$ of a thin layer has been proposed and demonstrated using only the reflection spectrum and taking into account the reflection of the rays from the free surface of the substrate. The accuracy of calculating the refractive index is about 0.2% and the absorption index about 1.5%.

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