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EXTRACTION OF OPTICAL PARAMETERS OF THIN FILMS FROM SPECTRAL MEASUREMENTS FOR DESIGN AND OPTICAL PERFORMANCE OF MULTILAYER STRUCTURES [*]

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Optical parameters of a-Si:H and indium tin oxide (ITO) thin films deposited on glass substrates are determined from spectral measurements of reflectance and/or transmittance. It is shown how important the exact knowledge of optical parameters as well as thicknesses of the layers for the design and the optical performance of multilayer structures is. The model of the p-i-n-based a:Si-H solar cell with ITO as transparent conductive oxide layer is used for illustrating. The modelling of the solar cell integral reflectance in the spectral region of (650–830) nm is used as a criterion to reverse engineering of a multilayer structure with suppressed reflectance losses. The reflectance of a solar cell is modelled and the simulation of the varying optical parameters of individual layers including their thicknesses is discussed. Besides this, the advantage of using an antireflective layer under ITO is discussed.

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1 Introduction

As optical properties of thin films differ markedly from those of the same bulk material and even are deposition-process-dependent, the knowledge of optical parameters of any individual thin film is central task for the design and performance of multilayer structures with thin films. Optical properties of hydrogenated amorphous silicon (a-Si:H) and indium tin oxide (ITO) that both often act in Schottky-type sensors (e.g. alignment and position sensors) and solar cells were determined from reflectance and/or transmittance spectra. Reflectance of a similar-to-real solar cell structure is modelled and discussed when varying optical parameters and antireflective coatings are used for the simulation. As light energy reflected from the entire structure is lost, from the optical point of view a solar cell should operate as a broad band-pass filter with suppressed reflectance losses.

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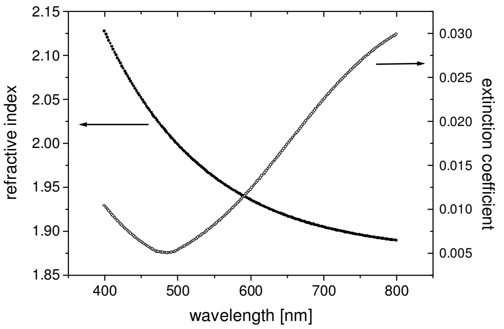

Fig. 1. n, k of ITO d = 105 nm.

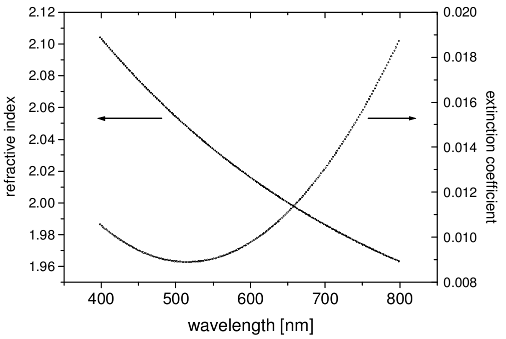

Fig. 2. n, k of ITO, d = 180 nm.

2 Optical properties of a single layer determined from spectral measurements

The classical physical optics describes the optical performance (reflectance R and/or transmittance T) of a single ideal parallel-sided homogeneous film with the average thickness d deposited on a thick incoherent substrate (with the thickness $\gg d$) illuminated at nearly normal incidence with monochromatic radiation. The film is characterised by the complex refractive index $N = (n - ik)$, where n is the real part of the complex refractive index, the imaginary part k is the extinction coefficient. The exact equations for R, T are in Appendix.

It is widely agreed that the optical parameters of thin films are deposition process-dependent. Therefore the knowledge of optical parameters of any individual thin film is necessary for the design and performance of any structure with thin films. Optical constants of the film can be deduced from the set of non-linear equations ("exp" refers to experimental values) at the wavelength λ

$$T(n,k,d,\lambda) - T_{\mathrm{exp}}(\lambda) = 0 \qquad R(n,k,d,\lambda) - R_{\mathrm{exp}}(\lambda) = 0\,. \tag{1}$$

While apparent interference effects are observed in the registered spectral region, then following equations must be fulfilled and can be used for evaluation of optical parameters

$$R_{\mathrm{max}}(n,k,d,\lambda) - R_{\mathrm{expmax}}(\lambda) = 0 \qquad R_{\mathrm{min}}(n,k,d,\lambda) - R_{\mathrm{expmin}}(\lambda) = 0 \tag{2}$$

$$T_{\mathrm{max}}(n,k,d,\lambda) - T_{\mathrm{expmax}}(\lambda) = 0 \qquad T_{\mathrm{min}}(n,k,d,\lambda) - T_{\mathrm{expmin}}(\lambda) = 0\,. \tag{3}$$

Equations (1)–(3) are non-linear in connection with the parameters of thin films involved and therefore, these can be extracted from equations (1)–(3) only by numerical methods. The optimisation procedures are often applied.

3 Experimental

ITO (indium tin oxide In_2O_3:SnO_2) is often used as transparent conductive oxide (TCO) in multilayer structures, e.g. solar cells, alignment or position sensors, electrochromic devices etc. ITO films under study (d approx. 170 nm, 300 nm) were deposited on cleaned microscope slides

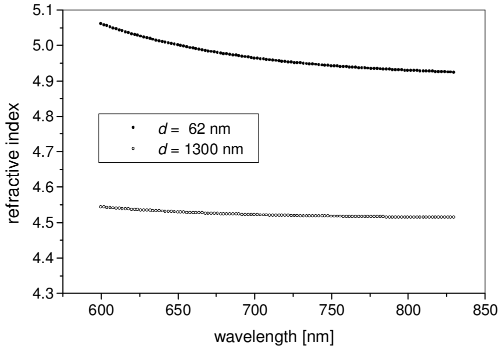

Fig. 3. Refractive index of a-Si:H.

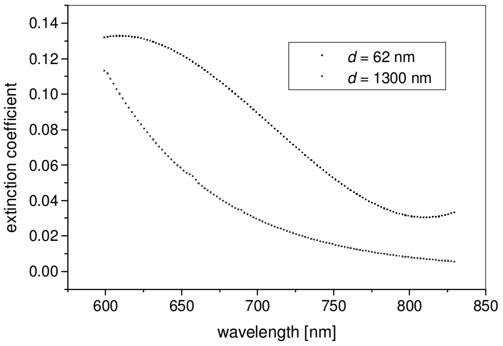

Fig. 4. Extinction coefficient of a-Si:H.

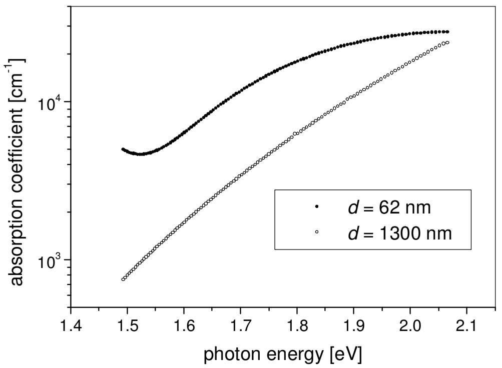

Fig. 5. Absorption coefficient of a-Si:H.

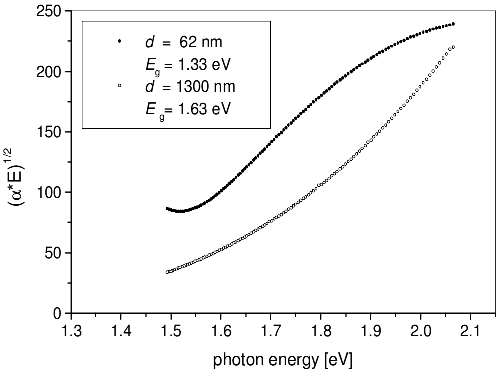

Fig. 6. Tauc plot of a-Si:H.

by diode sputtering by using the Perkin Elmer 2400/8L planar diode sputtering unit of rf power of 700 W from hot-pressed ceramic In_2O_3 target (9 mol % of SnO_2) in purity of 99.9%. During the deposition the substrate temperature was kept at 350°C and the total pressure of Ar working gas at 1.3 Pa. Microstructural analysis of ITO layer [1] showed the films being polycrystalline with average crystallite size of $\sim$ 30 nm.

Amorphous Si layers for solar cells of i-type were deposited on glass substrate in device quality by PECVD in rf hydrogen silane plasma at the temperature $\sim$ 240°C. Technological film thicknesses estimated from deposition conditions were $d \sim$ 50 nm, 950 nm.

Absolute spectral measurements of reflectance and transmittance at nearly normal incidence were carried out by the double-beam UV VIS Carl Zeiss Jena spectrophotometer Specord M40 with the 2.5 nm slit in the spectral region (400–900) nm at room temperature. Freshly evaporated aluminium sample in the reference beam was used for reflectance measurements. The absolute ordinate errors were ~ 0.01. Probed sample areas were ~ 0.2 cm^2. Spatial inhomogeneity was not investigated. The films were considered as homogeneous with parallel interfaces.

4 Results

Optical parameters of both ITO films and of the thinner a-Si:H film have been determined by *R/T* method according to the equations (1). Forouhi – Bloomer dispersion model was used with the genetic algorithm, no dispersion model but direct point-to-point method with the other numerical methods. Levenberg – Marquardt non-linear least squares method and genetic algorithm [2] were used to minimise $\sum_i [R_{\exp i} - R_i(\lambda_i, n, k, d)]^2$. As the thicker a-Si:H film reflectance/transmittance spectra manifest apparent interference effects, besides the genetic algorithm, the envelope method [3] was also used for retrieving the optical parameters.

As for a-Si:H solar cells only the spectral region of the light trapping is interesting, calculations were performed at wavelengths over $\sim$ 600 nm. The optimisation procedure was subjected to physical constraints considering the nature of n being decreasing function of λ and convex in the spectral region of interest. Results yield the optically determined ITO film thicknesses d = 105 nm, d = 180 nm, and the optical parameters in Figs. 1, 2. The results for a-Si:H thin films including Tauc optical band-gaps are in Figs. 3–6.

The results confirm the fact of n, k of thin films being deposition and thickness dependent. This can be markedly seen for a-Si:H samples, where d differs to a greater extent than for ITO samples.

5 Optical modelling of a solar cell with varying optical parameters and thickness

In a multilayer structure, optical properties of each individual layer manifest themselves in a complicated function of spectral reflectance $R(\lambda)$ or transmittance $T(\lambda)$ of the entire structure. Our model of a solar cell optical performance considers incoherent multiple reflections within non-absorbing slight-dispersive glass substrate (Corning 1737) as well as coherent reflections within all other layers assumed to be absorbing. The spectral reflectance $R(\lambda)$ of a solar cell in Fig. 7 including p-i-n-structure of a-Si:H has been simulated with realistic optical parameters [4,5] by a recursive algorithm with propagation of reflection responses (see e.g. [6]). As opposed to optical modelling in [7], the back metal layer under p-i-n structure has not been incorporated into our model. The thickness and the optical parameters of p-type and n-type a-Si:H have been kept constant.

The thickness and the optical parameters of ITO and the intrinsic a-Si:H layer have been varied. Some results of reflectance simulations are in Figs. 8–10. For the wavelengths below $\sim$ 550 nm with strong absorption of light in the structure there are small differences between spectral reflectances of solar cells with various optical parameters and layer thicknesses. For higher wavelengths, apparent interference in reflectance spectra occurs. Changing optical parameters and thicknesses are reflected in magnitudes and positions of interference fringes.

The integral reflectance $R_{\mathrm{int}} = \int_{650}^{830} R(\lambda)d\lambda$ over the spectral range of interest has been calculated to represent optical losses caused by the entire modelled structure. As $R(\lambda)$ and R_{int} as functions of optical parameters and d are periodical, the layer thickness d and the optical parameters could be adjusted to minimise the reflectance losses. Figs. 11–12 imply that R_{int} can be reduced with the variation of d and n of intrinsic layer by over 10 %. For the intrinsic layer, minimal R_{int} has been estimated at d $\sim$ 540 nm and with the spectral refractive index slightly

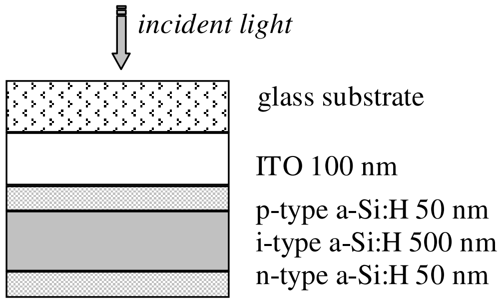

Fig. 7. Modelled solar cell structure.

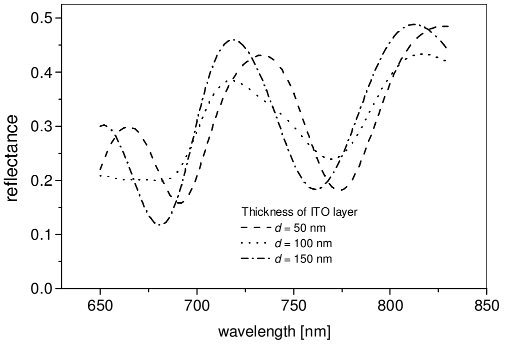

Fig. 8. The ITO layer thickness variation.

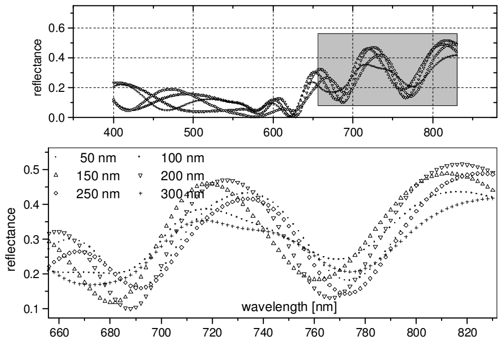

Fig. 9. Simulated reflectance of a-Si:H solar cell: the thicknesses of intrinsic layer variation.

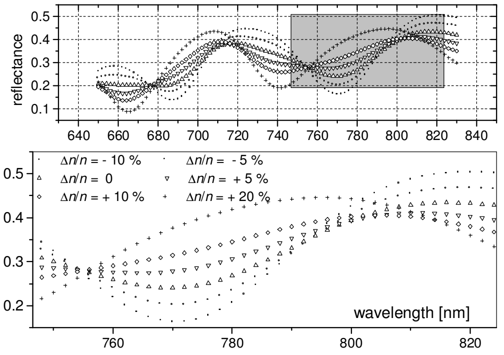

Fig. 10. Simulated reflectance of a-Si:H solar cell: the intrinsic layer refractive index variation.

below the assumed values. The ITO thicknesses best for the minimal R_{int} at $\lambda > 650$ nm are $d \sim 100$ nm, $d \sim 300$ nm (Fig. 13). Integral reflectance losses with $d = 300$ nm are lower by ~ 10 %. The choice between both thicknesses should be based on the better electrical performance of the TCO layer. The influence of the ITO extinction coefficient variations has been found not to be significant.

6 Modelling of a solar cell with reflectance losses reduced by AR coating

It has been reported that textured front reflector [8] can achieve the reduction of reflectance losses. Another contribution to the optical gains can be associated with microscopic roughness [9]. Modelling of the enhancement of the optical collections without texture has been performed in this work by various antireflective (AR) coatings with various d imbedded between various layers of the solar cell. Although it is not always easy to find materials with suitable refractive index dispersion, graded-index multilayer broad-band AR coatings are reported to perform well in optoelectronic devices [10]. In a real thin-film solar cell, besides the optical performance also

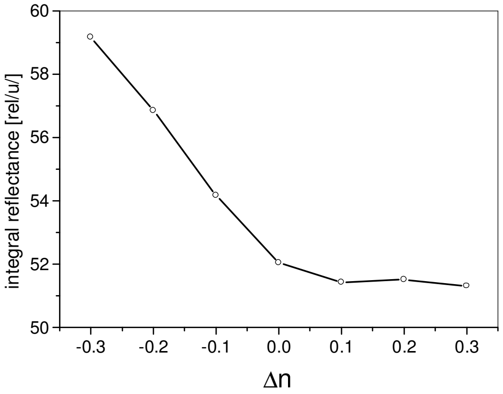

Fig. 11. Integral reflectance versus the intrinsic layer refractive index.

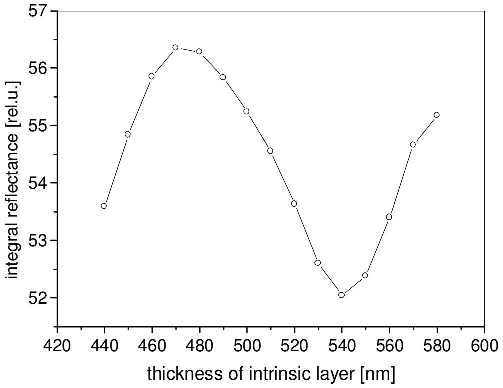

Fig. 12. Integral reflectance versus the intrinsic layer thickness variation.

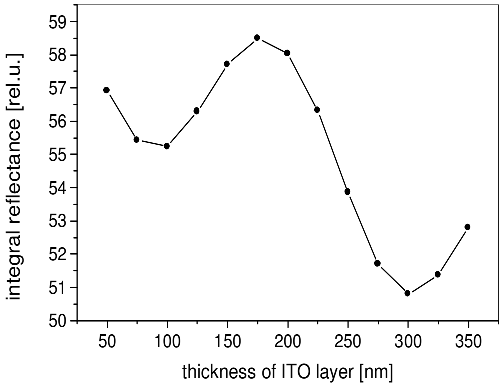

Fig. 13. Integral reflectance versus the thickness of ITO layer.

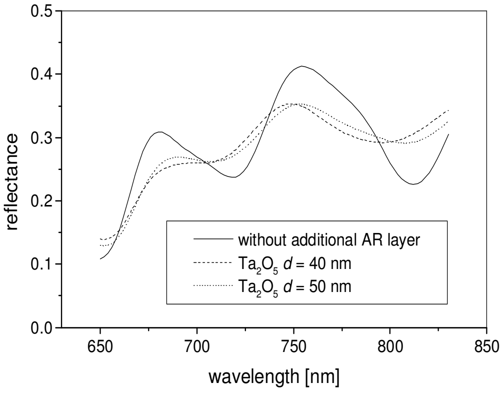

Fig. 14. Solar cell with additional AR coating between ITO and p-type a-Si:H.

other specifications such as quantum efficiency and stability are of vital importance. For the reduction of optical losses by AR coatings, regard to production cost and engineering problems should contribute to a tolerant configuration.

Owing to its high refractive index and high transparency, ITO layer sandwiched between glass and p-i-n a-Si:H structure could act quite well as an natural single AR coating of the non-quarter thickness. The simulations have proved that detuning the ITO thickness from the quarter-wave value broadens the spectral range of minimised reflectances.

Another non-quarter-wave AR coating imbedded between ITO and a-Si:H requires the refractive index $n_{\mathrm{AR}} = \sqrt{n_{\mathrm{ITO}} n_{\mathrm{p-type\ a-Si:H}}} \sim 2.3$ (assumed to be non-absorbing and non-dispersive in this approximation) for (400–800) nm. It should be noted that this structure acts as non-quarter—non-quarter double AR coating owing to the requirement of wide spectral response. Appropriate candidates (with refractive indices in parentheses at $\lambda \sim 700$ nm) seem to be e.g. ZnS (2.36), Ta_2O_5 (2.25), TiO_2 (2.36), CeO_2 (2.35), Nb_2O_5 (2.29). The simula-

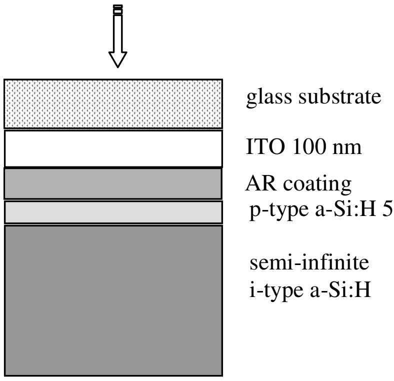

Fig. 15. Structure under modelling of passive performance.

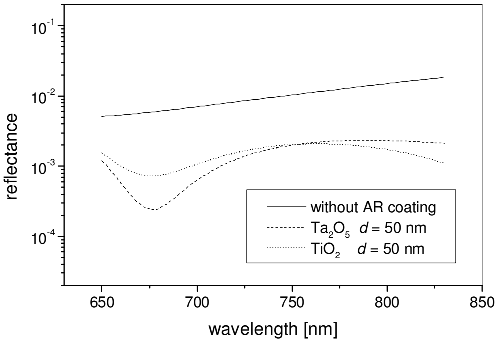

Fig. 16. Passive version of reflectance with additional AR coating.

tion was performed with the above materials with spectral refractive indices taken, e.g. from [11] and negligible extinction coefficients (except for TiO_2). Our AR coatings policy was focused on the spectral reflectance not to be necessarily equal to zero as it matched the criterion $\int_\lambda R_{\text{without AR}}(\lambda)\, d\lambda > \int_\lambda R_{\text{with AR}}(\lambda) d\lambda$. The optical modelling showed that further reduction of integral reflectance in the region (650–830) nm could be achieved. The demonstration is in Fig. 14 for Ta_2O_5. The best result with the R_{int} reduction by $\sim 5\%$ was simulated with Ta_2O_5 of d = 50 nm.

We have also modelled the multilayer performance with additional Ta_2O_5 ($d = 50$ nm) AR coating as seen at the interface of p and i-type a-Si:H with semi-infinite intrinsic layer (Fig. 15). This performance can be considered as passive while the optical performance of the entire solar cell as active. The spectral reflectance in the entire spectral region of interest (Fig. 16) drops well below 10^{-2} and $R_{\text{int}} = 0.27$ (the same value of R_{int} was achieved with TiO_2, $d = 30$ nm), while without AR $R_{\text{int}} = 1.85$.

7 Conclusions

Optical parameters of ITO and a-Si:H layers entering the solar cell structure have been determined by R/T method. Simulations of solar cell optical performances have been performed. They show that precise control of optical parameters and film thicknesses is advantageous and that over 10 % more of incident light energy can be directed into the light trapping intrinsic layer when the appropriate layer thicknesses and optical parameters are chosen. This confirms the importance of the reverse engineering of the solar cell design and the importance of the exact optical characterization of all individual thin films. Concept of the solar cell design should include theoretical design with required specifications (application of an optimisation technique, e.g. genetic algorithm is recommended), the determination of each single layer optical data from ex-situ measurements and the correlation of the multilayer spectrum calculated with measured data with the required theoretical design.

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Appendix

Reflectance of a single layer with the thickness d on thick substrate is given by the expression [e.g. 3,6,12]

$$R = \frac{A + Bx + Cx^2}{D + Ex + Fx^2}\,,$$

where $x = \exp\left(-\alpha d\right)$ is the absorbance, $\alpha = (4\pi k_1)/\lambda$ is the absorption coefficient and

$$\begin{aligned}
A &= \left[\left(1-n_1\right)^2 + k_1^2\right]\left[\left(n_1+n_2\right)^2 + \left(k_1+k_2\right)^2\right]\\
C &= \left[\left(1-n_1\right)^2 + k_1^2\right]\left[\left(n_1-n_2\right)^2 + \left(k_1-k_2\right)^2\right]\\
D &= \left[\left(1+n_1\right)^2 + k_1^2\right]\left[\left(n_1+n_2\right)^2 + \left(k_1+k_2\right)^2\right]\\
F &= \left[\left(1-n_1\right)^2 + k_1^2\right]\left[\left(n_1-n_2\right)^2 + \left(k_1-k_2\right)^2\right]\\
B &= 2\left[A'\cos\varphi + B'\sin\varphi\right]\\
E &= 2\left[C'\cos\varphi + D'\sin\varphi\right]\\
A' &= \left(1+n_1^2-k_1^2\right)\left(n_1^2-n_2^2+k_1^2-k_2^2\right) + 4k_1\left(n_1k_2-n_2k_1\right)\\
C' &= \left(1-n_1^2-k_1^2\right)\left(n_1^2-n_2^2+k_1^2-k_2^2\right) - 4k_1\left(n_1k_2-n_2k_1\right)\\
B' &= 2\left(1+n_1^2-k_1^2\right)\left(n_1k_2-n_2k_1\right) - 2k_1\left(n_1^2-n_2^2+k_1^2-k_2^2\right)\\
D' &= 2\left(1-n_1^2-k_1^2\right)\left(n_1k_2-n_2k_1\right) + 2k_1\left(n_1^2-n_2^2+k_1^2-k_2^2\right)
\end{aligned}$$

$\varphi = (4\pi n_1 d)/\lambda$ is the phase factor, index “1” refers to the thin film, index “2” to the substrate.

Transmittance of a single layer with the thickness d on thick non-absorbing substrate is expressed by the equation

$$T = \frac{A}{B - Cx + Dx^2}\,,$$

where

$$\begin{aligned}
A &= 16n_2\left(n_1^2+k_1^2\right)\\
B &= \left[\left(1+n_1\right)^2 + k_1^2\right]\left[\left(n_1+n_2^2\right)\left(n_1+1\right) + k_1^2\right]\\
C &= 2\cos\varphi\left[\left(n_1^2+k_1^2-1\right)\left(n_1^2+k_1^2-n_2^2\right) - 2k_1^2\left(n_1^2+1\right)\right]\\
&\quad - 2\sin\varphi\left[\left(n_1^2+k_1^2-1\right)\left(n_2^2+1\right) + 2\left(n_1^2+k_1^2-n_2^2\right)\right]\\
D &= \left[\left(n_1-1\right)^2 + k_1^2\right]\left[\left(n_1-1\right)\left(n_1-n_2^2\right) + k_1^2\right]
\end{aligned}$$

and the absorbance $x = \exp\left(-\alpha d\right)$, the absorption coefficient $\alpha = (4\pi k_1)/\lambda$ and the phase shift $\varphi = (4\pi n_1 d)/\lambda$ are the same physical quantities as in the expression for R.

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