Estimation of the optical constants and the thickness of thin films using unconstrained optimization

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Abstract

The problem of estimating the thickness and the optical constants of thin films using transmission data only is very challenging from the mathematical point of view, and has a technological and an economic importance. In many cases it represents a very ill-conditioned inverse problem with many local-nonglobal solutions. In a recent publication we proposed nonlinear programming models for solving this problem. Well-known software for linearly constrained optimization was used with success for this purpose. In this paper we introduce an unconstrained formulation of the nonlinear programming model and we solve the estimation problem using a method based on repeated calls to a recently introduced unconstrained minimization algorithm. Numerical experiments on computer-generated films show that the new procedure is reliable.

1 Introduction

For most modern applications of thin dielectric or semiconductor films, the optical properties of interest cover a photon energy range around the fundamental absorption edge of the material. Moreover, as the applications make use of multiple coherent reflections at the interfaces, the thickness of the films is an important design and characterization parameter. Optical transmittance provides accurate and rapid information on the spectral range where the material goes from complete opacity to some degree of transparency [1,2]. As a consequence, the problem of retrieving the optical constants ($\widetilde{n}\ (\lambda) = n(\lambda) + ik(\lambda)$) and the thickness ($d$) of thin films, from transmission data only, is of particular importance. Some useful approximate solutions have been found in cases where the transmittance displays an interference pattern in a highly transparent spectral region [3,4]. Up to now, however, the general solution of the problem has been elusive, because the system of equations is highly underdetermined. Recently, we reported a new method, based on a pointwise constrained optimization approach, which allows to solve the general case [5,6]. The method defines a nonlinear programming problem, the unknowns of which are the coefficients to be estimated, with linear constraints that represent prior knowledge about the physical solution. The retrieval of the correct thickness and optical constants of the films does not rely on the existence of interference fringes. The new method was successful in retrieving d and $\widetilde{n}\ (\lambda)$ from very different transmission spectra of computer made and real world films [5,6]. The main inconvenient of the pointwise constrained optimization approach [5,6] is that is a rather complex large-scale linearly constrained nonlinear programming problem whose solution can be obtained only by means of rather sophisticated and not always available computer codes that can deal effectively with sparsity of the matrix

of constraints [7,8].

We consider then the problem of estimating the absorption coefficients, the refractive index and the thickness of thin films, using transmission data only. Given the wavelength λ, the refractive index of the substrate s and the unknowns d (thickness), $n(\lambda)$ (refractive index) and $k(\lambda)$ (attenuation coefficient), the theoretical transmission is given by a well-known formula [2,4]. Having measurements of the transmission at (many) different wavelengths we want to estimate the above mentioned unknowns. At a first glance, this problem is highly underdetermined since, for each wavelength, the single equation

$$Theoretical\ transmission\ = Measured\ transmission \tag{1}$$

has three unknowns $d, n(\lambda), k(\lambda)$ and only d is repeated for all values of λ. The driving idea in [5,6] was to incorporate prior knowledge on the functions $n(\lambda), k(\lambda)$ in order to decrease the degrees of freedom of (1) up to a point that only physically meaningful estimated parameters are admitted.

The idea of assuming a closed formula for n and k depending of few coefficients has been already reported [3,4]. The methods originating from this idea are efficient when the transmission curve exhibits a fringe pattern representing rather large spectral zones were $k(\lambda)$ is almost null. In other cases, the satisfaction of (1) is very rough or the curves $n(\lambda),\ k(\lambda)$ are physically unacceptable.

In [5,6], instead of imposing a functional form to $n(\lambda)$ and $k(\lambda)$, the phenomenological constraints that restrict the variability of these functions were stated explicitly so that the estimation problem took the form:

$$Minimize \sum_{\lambda} [Theoretical\ transmission\ (\lambda) - Measured\ transmission\ (\lambda)]^2$$
$$subject\ to\ \textbf{Physical Constraints}. \tag{2}$$

In this way, well behaved functions $n(\lambda)$ and $k(\lambda)$ can be obtained without severe restrictions that may damage the quality of the fitting (1).

The main contribution of the present paper is to establish a method for solving the estimation problem where (2) is replaced by an unconstrained optimization problem. We solved this problem using a very simple algorithm introduced recently by Raydan [9]. This method realizes a very effective idea for potentially large-scale unconstrained minimization. It consists of using only gradient directions with steplengths that ensure rapid convergence. The reduction of (2) to an unconstrained minimization problem needed the calculation of very complicate derivatives of functions, which could not be possible without the use of automatic differentiation techniques. Here we used the procedures for automatic differentiation described in [10].

2 Unconstrained formulation of the estimation problem

The transmission T of a thin absorbing film deposited on a thick transparent substrate (see [4]) is given by:

$$T = \frac{Ax}{B - Cx + Dx^2} \tag{3}$$

where

$$A = 16s(n^2 + k^2) \tag{4}$$

$$B = [(n+1)^2 + k^2][(n+1)(n+s^2) + k^2] \tag{5}$$

$$\begin{aligned} C = {} & [(n^2 - 1 + k^2)(n^2 - s^2 + k^2) - 2k^2(s^2 + 1)]2\cos\varphi \\ & -k[2(n^2 - s^2 + k^2) + (s^2 + 1)(n^2 - 1 + k^2)]2\sin\varphi \end{aligned} \tag{6}$$

$$D = [(n-1)^2 + k^2][(n-1)(n-s^2) + k^2] \tag{7}$$

$$\varphi = 4\pi nd/\lambda, \quad x = exp(-\alpha d), \quad \alpha = 4\pi k/\lambda. \tag{8}$$

In formulae (4)–(8) the following notation is used:

(a) λ is the wavelength;

(b) $s = s(\lambda)$ is the refractive index of the transparent substrate (assumed to be known),

(c) $n = n(\lambda)$ is the refractive index of the film;

(d) $k = k(\lambda)$ is the attenuation coefficient of the film (α is the absorption coefficient);

(e) d is the thickness of the film.

A set of experimental data $(\lambda_i, T^{meas}(\lambda_i))$, $\lambda_{min} \leq \lambda_i < \lambda_{i+1} \leq \lambda_{max}$, for $i = 1, \ldots, N$, is given, and we want to estimate d, $n(\lambda)$ and $k(\lambda)$. This problem seems highly underdetermined. In fact, for known d and given λ, the following equation must hold:

$$T(\lambda, s(\lambda), d, n(\lambda), k(\lambda)) = T^{meas}(\lambda). \tag{9}$$

Equation (9) has two unknowns $n(\lambda)$ and $k(\lambda)$ and, therefore, in general, its set of solutions is a curve in the two-dimensional $(n(\lambda), k(\lambda))$ space. Therefore, the set of functions (n, k) that satisfy (9) for a given d is infinite and, roughly speaking, is represented by a nonlinear manifold of dimension N in $\mathbb{R}^{2N}$.

However, physical constraints reduce drastically the range of variability of the unknowns $n(\lambda), k(\lambda)$. For example, in the neighborhood of the fundamental absorption edge (normal dispersion), these physical constraints are:

PC1: $n(\lambda) \geq 1, \;\; k(\lambda) \geq 0 \quad$ for all $\;\lambda \in [\lambda_{min}, \lambda_{max}]$;

PC2: $n(\lambda)$ and $k(\lambda)$ are decreasing functions of λ;

PC3: $n(\lambda)$ is convex;

PC4: There exists $\lambda_{infl} \in [\lambda_{min}, \lambda_{max}]$ such that $k(\lambda)$ is convex if $\lambda \geq \lambda_{infl}$ and concave if $\lambda < \lambda_{infl}$.

Observe that, assuming **PC2**, **PC1** is satisfied under the sole assumption $n(\lambda_{max}) \geq 1$ and $k(\lambda_{max}) \geq 0$. The constraints **PC2**, **PC3** and **PC4** can be written, respectively, as

$$n'(\lambda) \leq 0, \quad k'(\lambda) \leq 0 \text{ for all } \lambda \in [\lambda_{min}, \lambda_{max}]; \tag{10}$$

$$n''(\lambda) \geq 0 \text{ for all } \lambda \in [\lambda_{min}, \lambda_{max}]; \tag{11}$$

$$k''(\lambda) \leq 0 \text{ for } \lambda \in [\lambda_{min}, \lambda_{infl}] \text{ and} \tag{12}$$

$$k''(\lambda) \geq 0 \text{ for } \lambda \in [\lambda_{infl}, \lambda_{max}]. \tag{13}$$

Clearly, the constraints

$$n''(\lambda) \geq 0 \text{ for all } \lambda \in [\lambda_{min}, \lambda_{max}] \text{ and } n'(\lambda_{max}) \leq 0$$

imply that

$$n'(\lambda) \leq 0 \quad \text{for all} \quad \lambda \in [\lambda_{min}, \lambda_{max}].$$

Moreover,

$$k''(\lambda) \geq 0 \quad \text{for all} \quad \lambda \in [\lambda_{infl}, \lambda_{max}] \text{ and } k'(\lambda_{max}) \leq 0$$

imply that

$$k'(\lambda) \leq 0 \quad \text{for all} \quad \lambda \in [\lambda_{infl}, \lambda_{max}].$$

Finally,

$$k''(\lambda) \leq 0 \quad \text{for all} \quad \lambda \in [\lambda_{min}, \lambda_{infl}] \ \text{ and } \ k'(\lambda_{min}) \leq 0$$

imply that

$$k'(\lambda) \leq 0 \quad \text{for all} \quad \lambda \in [\lambda_{min}, \lambda_{infl}].$$

Therefore, **PC2** can be replaced by

$$n'(\lambda_{max}) \leq 0, \quad k'(\lambda_{max}) \leq 0, \quad k'(\lambda_{min}) \leq 0. \tag{14}$$

Summing up, the assumptions **PC1**–**PC4** will be satisfied if, and only if,

$$n(\lambda_{max}) \geq 1, \quad k(\lambda_{max}) \geq 0, \tag{15}$$

$$n'(\lambda_{max}) \leq 0, \quad k'(\lambda_{max}) \leq 0, \tag{16}$$

$$n''(\lambda) \geq 0 \ \text{ for all } \ \lambda \in [\lambda_{min}, \lambda_{max}], \tag{17}$$

$$k''(\lambda) \geq 0 \ \text{ for all } \ \lambda \in [\lambda_{infl}, \lambda_{max}], \tag{18}$$

$$k''(\lambda) \leq 0 \ \text{ for all } \ \lambda \in [\lambda_{min}, \lambda_{infl}], \tag{19}$$

$$k'(\lambda_{min}) \leq 0. \tag{20}$$

So, the continuous least squares solution of the estimation problem is the solution $(d, n(\lambda), k(\lambda))$ of

$$\text{Minimize} \quad \int_{\lambda_{min}}^{\lambda_{max}} |T(\lambda, s(\lambda), d, n(\lambda), k(\lambda)) - T^{meas}(\lambda)|^2 d\lambda \tag{21}$$

subject to the constraints (15)–(20).

Our idea in this work is to eliminate, as far as possible, the constraints of the problem, by means of a suitable change of variables. Roughly speaking,

we are going to put the objective function (21) as depending on the second derivatives of $n(\lambda)$ and $k(\lambda)$ plus functional values and first derivatives at λ_{max}. Moreover, positivity will be guaranteed expressing the variables as squares of auxiliary unknowns. In fact, we write

$$n(\lambda_{max}) = 1 + u^2, \quad k(\lambda_{max}) = v^2, \tag{22}$$

$$n'(\lambda_{max}) = -u_1^2, \quad k'(\lambda_{max}) = -v_1^2, \tag{23}$$

$$n''(\lambda) = w(\lambda)^2 \ \text{ for all } \lambda \in [\lambda_{min}, \lambda_{max}], \tag{24}$$

$$k''(\lambda) = z(\lambda)^2 \ \text{ for all } \lambda \in [\lambda_{infl}, \lambda_{max}], \tag{25}$$

$$k''(\lambda) = -z(\lambda)^2 \ \text{ for all } \lambda \in [\lambda_{min}, \lambda_{infl}]. \tag{26}$$

At this point, in order to avoid a rather pedantic continuous formulation of the problem, we consider the real-life situation, in which data are given for a set of N equally spaced points on the interval $[\lambda_{min}, \lambda_{max}]$. So, we define

$$h = (\lambda_{max} - \lambda_{min})/(N-1),$$

$$\lambda_i = \lambda_{min} + (i-1)h, \quad i = 1, \ldots, N.$$

Consequently, the measured transmission at λ_i will be called T_i^{meas}. Moreover, we will use the notation n_i, k_i, w_i, z_i for the finite difference estimates of $n(\lambda_i)$, $k(\lambda_i)$, $w(\lambda_i)$ and $z(\lambda_i)$:

$$n_i \approx n(\lambda_i), \quad k_i \approx k(\lambda_i),$$

$$w_i \approx w(\lambda_{i+1}), \quad z_i \approx z(\lambda_{i+1}),$$

for all $i = 1, \ldots, N$. Discretization of the differential relations (22-26) gives:

$$n_N = 1 + u^2, \quad v_N = v^2, \tag{27}$$

$$n_{N-1} = n_N + u_1^2 h, \quad k_{N-1} = k_N + v_1^2 h, \tag{28}$$

$$n_i = w_i^2 h^2 + 2n_{i+1} - n_{i+2}, \quad i = 1, \ldots, N-2, \tag{29}$$

$$k_i = z_i^2 h^2 + 2k_{i+1} - k_{i+2} \ \text{ if } \ \lambda_{i+1} \geq \lambda_{infl}, \tag{30}$$

$$k_i = -z_i^2 h^2 + 2k_{i+1} - k_{i+2} \ \text{ if } \ \lambda_{i+1} < \lambda_{infl}. \tag{31}$$

Finally, the objective function (21) is approximated by a sum of squares, giving the optimization problem

$$\text{Minimize} \quad \sum_{i=1}^{N} [T(\lambda_i, s(\lambda_i), d, n_i, k_i) - T_i^{meas}]^2 \tag{32}$$

subject to

$$k_1 \geq k_2 \tag{33}$$

Since n_i and k_i depend on u, u_1, v, v_1, w, z and λ_{infl} through (27–31), problem (32) takes the form

$$\text{Minimize} \quad f(d, \lambda_{infl}, u, u_1, v, v_1, w_1, \ldots, w_{N-2}, z_1, \ldots, z_{N-2})) \tag{34}$$

subject to (33).

We expect that the constraint (33) will be inactive at a solution of (34-33), so that we are going to consider the unconstrained problem (34). The unknowns that appear in (34) have a different nature. The thickness d is a dimensional variable (measured in nanometers in our real-life problems) that can be determined using the observations $T^{meas}(\lambda_i)$ for (say) $\lambda_i \geq \lambda_{bound}$, where λ_{bound}, an upper bound for λ_{infl}, reflects our prior knowledge of the problem. For this reason, our first step in the estimation procedure will be to estimate d using data that correspond to $\lambda_i \geq \lambda_{bound}$. For accomplishing this objective we solve the problem

$$\text{Minimize} \quad \bar{f}(u, u_1, v, v_1, w, z) \equiv \sum_{\lambda_i \geq \lambda_{bound}} [T(\lambda_i, s(\lambda_i), d, n_i, k_i) - T_i^{meas}]^2 \tag{35}$$

for different values of d and we take as estimated thickness the one that gives the lowest functional value. In this case the constraint (33) is irrelevant since it is automatically satisfied by the convexity of k and the fact that the derivative of k and λ_{min} are nonpositive. From now on we consider that d is fixed, coming from the procedure above.

The second step consists of determining λ_{infl}, together with the unknowns u, u_1, v, v_1, w, z. For this purpose observe that, given d and λ_{infl} the problem

$$\text{Minimize } \sum_{i=1}^{N} [T(\lambda_i, s(\lambda_i), d, n_i, k_i) - T_i^{meas}]^2 \tag{36}$$

is (neglecting (33)) an unconstrained minimization problem whose variables are u, u_1, v, v_1, w, z ($2N$ variables). We solve this problem for several trial values of λ_{infl} and we take as estimates of n and k the combination of variables that gives the lowest value. For minimizing this function and for solving (35) for different trial thickness, we use the unconstrained minimization solver that will be described in the next section.

3 Description of the unconstrained minimization algorithm

As we saw in the previous section, the unconstrained minimization problem (35) and (36) have the form

$$\text{Minimize} \quad f(u, u_1, v, v_1, w_1, \ldots, w_{N-2}, z_1, \ldots, z_{N-2}). \tag{37}$$

In order to simplify the notation, in this section we will write

$$\mathbf{x} = (u, u_1, v, v_1, w_1, \ldots, w_{N-2}, z_1, \ldots, z_{N-2}).$$

Partial derivatives of f are usually necessary in optimization algorithms, since they provide the first-order information on the objective function that allows computational algorithms to follow downhill trajectories. In this case, derivatives are very hard to compute. For this reason it was necessary to use an automatic differentiation procedure (reverse mode) for performing this task. See [10] for details.

In principle, any unconstrained optimization algorithm can be used to solve (37) (see [11-13]). Since the problem has, potentially, a large number of variables, our choice must be restricted to methods that are able to cope with that situation. A recent paper by Raydan [9] induced us to use the Spectral Gradient Method (SGM), an implementation of the Barzilai-Borwein for quadratics introduced in [9]. In fact, Raydan showed, using a well known set of classical test problems, that SGM outperforms conjugate gradient algorithms (see [12,13]) for large scale unconstrained optimization. Raydan's spectral gradient method is extremely easy to implement, a fact that contributed to support our decision, since it enables us to become independent of black-box like imported software. Our description of SGM here is, essentially, the one of Raydan with a small difference in the choice of the step α_k when $b_k \leq 0$.

We denote $g(\mathbf{x}) = \nabla f(\mathbf{x})$. The algorithm starts with $\mathbf{x}_0 \in I\!R^n$ and uses an integer $M \geq 0$, a small parameter $\varepsilon > 0$, a sufficient decrease parameter $\gamma \in (0,1)$ and safeguarding parameters $0 < \sigma_1, < \sigma_2 < 1$. Initially, $\alpha_0 \in [1/\varepsilon, \varepsilon]$ is arbitrary. Given $\mathbf{x}^k \in I\!R^n$, $\alpha_k \in [1/\varepsilon, \varepsilon]$, Algorithm 3.1 describes how to obtain $\mathbf{x}_{k+1}$ and α_{k+1}, and when to terminate the process.

Algorithm 3.1

Step 1. *Detect whether the current point is stationary*

If $\|g(\mathbf{x}^k))\| = 0$, terminate the generation of the sequence, declaring that $\mathbf{x}^k$ is stationary.

Step 2. *Backtracking*

Step 2.1 Set $\lambda \leftarrow \alpha_k$.

Step 2.2 Set $\mathbf{x}^+ = \mathbf{x}^k - \lambda g(\mathbf{x}^k)$.

Step 2.3 If

$$f(\mathbf{x}^+) \leq \max_{j \geq \min\{0,k-M\}} f(\mathbf{x}^j) + \gamma \langle \mathbf{x}^+ - \mathbf{x}^k, g(\mathbf{x}^k) \rangle, \tag{38}$$

then define $\mathbf{x}^{k+1} = \mathbf{x}^k$, $\mathbf{s}^k = \mathbf{x}^{k+1} - \mathbf{x}^k$, $\mathbf{y}^k = g(\mathbf{x}^{k+1}) - g(\mathbf{x}^k)$ and go to Step 3.

If (38) does not hold, define

$$\lambda_{new} \in [\sigma_1 \lambda, \sigma_2 \lambda], \tag{39}$$

set $\lambda \leftarrow \lambda_{new}$ and go to Step 2.2.

Step 3 *Compute spectral steplenght*

Compute $b_k = \langle \mathbf{s}^k, \mathbf{y}^k \rangle$.

If $b_k \leq 0$, set $\alpha_{k+1} = 1/\varepsilon$, else, compute $a_k = -\langle \mathbf{s}^k, g(\mathbf{x}^k) \rangle$ and

$$\alpha_{k+1} = \min \{1/\varepsilon, \max \{\varepsilon, a_k/b_k\}\}.$$

In practice the computation of λ_{new} uses one-dimensional quadratic interpolation and it is safeguarded with (39).

4 Numerical results

In order to test the reliability of the new unconstrained optimization approach we used the computer-generated transmission of *gedanken* films deposited onto glass or crystalline silicon substrates. In the simulations the refractive index of the glass $s_{glass}(\lambda)$ is given by:

$$s_{glass}(\lambda) = \sqrt{1 + 1/(0.76194 - 7940/\lambda^2)},$$

and the refractive index of the silicon substrate $s_{Si}(\lambda)$ was assumed to be given by:

$$s_{Si}(\lambda) = 3.71382 - 8.6912310^{-5}\lambda - 2.4712510^{-8}\lambda^2 + 1.0467710^{-11}\lambda^3.$$

In all the simulations, we assume that the wavelength and the thickness are measured in nanometers. The transmission $T^{meas}(\lambda)$ for each film was first computed in the range $\lambda \in [\lambda_{\min}, \lambda_{\max}]$ using a known thickness d_{true} and a known refractive index $n_{true}(\lambda)$ and absorption coefficient $\alpha_{true}(\lambda)$. In order to consider realistic situations, including experimental inaccuracy, we considered alternative computations of $T^{meas}(\lambda)$, were the true transmission was rounded to four, three and two decimals. We also performed numerical experiments using a different number of transmission points: 100, 50 and 25. The description of these gedanken films and the corresponding numerical results are given below.

Film A. This computer-generated film simulates an amorphous germanium thin film deposited on a glass substrate with $d_{true} = 118$ nm. The computed transmission $T^{meas}(\lambda)$ [$\lambda_{\min} = 600$ nm, $\lambda_{\max} = 2000$ nm],the functions $n_{true}(\lambda)$ and $\alpha_{true}(\lambda)$ are shown as continuous lines in Fig. 1. Note that $n_{true}(\lambda)$ and $\alpha_{true}(\lambda)$ are represented as a function of photon energy.

Film B. This computer-generated film is identical to **Film A** except for its thickness $d_{true} = 782$ nm. The *true* values of T^{meas}[$\lambda_{\min} = 1000$ nm, $\lambda_{\max} = 2000$ nm], $n(\lambda)$, and $\alpha(\lambda)$ are shown in Fig.2.

Film C. This computer-generated film simulates an amorphous germanium thin film deposited on a crystalline silicon substrate with $d_{true} = 147$ nm, $\lambda_{\min} = 1250$ nm and $\lambda_{\max} = 2500$ nm. The computed transmission $T^{meas}(\lambda)$ and the functions $n_{true}(\lambda)$ and $\alpha_{true}(\lambda)$ are shown as continuous lines in Fig. 3. Note that, again, $n_{true}(\lambda)$ and $\alpha_{true}(\lambda)$ are represented as a function of photon energy.

Film D. This is also a simulated film of amorphous germanium over a c-Si substrate. A thickness $d_{true} = 640$ nm has been assumed. The transmission was computed in the [$\lambda_{\min} = 640$ nm, $\lambda_{\max} = 1250$ nm] interval. Figure 3 shows, as continuous lines the computed values.

Film E. Simulated film of hydrogenated amorphous silicon deposited onto glass with $d_{true} = 624$ nm, T^{meas} [$\lambda_{\min} = 600$ nm, $\lambda_{\max} = 1600$ nm]. See also n_{true} and α_{true} in Fig. 5.

For our calculations we need initial estimates of $k(\lambda)$ and $n(\lambda)$. As initial estimate of $k(\lambda)$ we used a piecewise linear function the values of which are 0.1 at the smallest wavelength of the spectrum, 0.01 at $\lambda_{min}+0.2(\lambda_{max}-\lambda_{min})$ and 10^{-10} at λ_{max}. The initial estimate of $n(\lambda)$ is a linear function varying between 5 ($\lambda_{\min}$) and 3 ($\lambda_{\max}$) with step 1 (these values were chosen because of the previous knowledge of the simulated materials). As we exclude the constant functions, i.e., linear functions with the left extreme equal to the right one, for which preliminary tests showed us they lead the method to local minimizers, we have three possibilities for the initial estimate of $n(\lambda)$: the decreasing linear functions defined by the pairs of points $[(\lambda_{min}, 4); (\lambda_{max}, 3)]$, $[(\lambda_{min}, 5); (\lambda_{max}, 3)]$ and $[(\lambda_{min}, 5); (\lambda_{max}, 4)]$.

The general scheme to obtain the optimal parameters of these films is as follows. First, we need to break down the spectrum into two parts: $[\lambda_{min}, \lambda_{bound}]$ and $[\lambda_{bound}, \lambda_{max}]$, where λ_{bound} is a known upper bound of λ_{infl}. To estimate the thickness we use the points with abcissa belonging to $[\lambda_{bound}, \lambda_{max}]$. The procedure consist in running Algorithm 3.1 for different values of d between $d_{min} = \frac{1}{2}d_{kick}$ and $d_{max} = \frac{3}{2}d_{kick}$ with step 10 ($d_{\min},\ d_{\min} + 10,\ d_{\min} + 20, \ldots$), where d_{kick} can be a rough initial estimate of the true thickness. In this way, we obtain d_{trial}, the thickness value for which the smallest quadratic error occurs. Then we repeat the procedure

with $d_{\min} = d_{trial} - 10$, $d_{\max} = d_{trial} + 10$ and step 1 obtaining, finally, the estimated thickness d_{best}.

To estimate the inflection point we proceed in an analogous way, using the whole spectrum and the thickness fixed at d_{best}, trying different possible inflection points (obviously between $\lambda_{\min}$ and λ_{bound}) and taking as estimated inflection point the one which gives the smallest quadratic error. In all the runs just described, we allow only 3000 iterations of Algorithm 3.1. The final step of the method consists on fixing d_{best} and $\lambda_{\inf\, l}$ and running Algorithm 3.1 once more allowing 30000 iterations.

All the experiments were run in a SPARCstation Sun Ultra 1, with an UltraSPARC 64 bits processor, 167-MHz clock and 128-MBytes of RAM memory. We used the language C++ with the g++ compiler (GNU project C and C++ compiler v2.7) and the optimization compiler option -O4. In spite of the many executions of the unconstrained minimization algorithm that are necessary to solve each problem, the total CPU time used under the mentioned computer environment for the complete process never exceeded 10 minutes.

Table I corresponds to **Film A** only. It shows the precision obtained in $n(\lambda)$ and $\alpha(\lambda)$ using 25, 50 and 100 measured transmission points, and rounding the transmission data to two, three and four decimal places after the decimal point and, finally, without rounding. The errors reported are the maximum values of $\mid n(\lambda) - n_{true}(\lambda) \mid$ and $\mid \alpha(\lambda) - \alpha_{true}(\lambda) \mid$ for large and small photon energy spectral regions, respectively. Table II corresponds to the same film. It shows the estimated thickness for 25, 50 and 100 data points and different number of decimal places in $T^{meas}(\lambda)$. Table III shows the estimated thickness, and the quadratic errors obtained in the minimization process.

Figures 1 to 5 are self explanatory. Continuous lines are true transmissions, true refractive indices and true absorption coefficients for the five considered examples. The open circles represent best estimates (using all decimal places). Finally, Figs. 6, 7 and 8 show how the process of estimating the thickness worked for each different film.

5 Conclusions

The analysis of the numerical results allow us to draw the following conclusions:

1) The proposed procedure is highly reliable for estimating the true thickness in all films when four (or all) digits of the transmission data are used. The method provides a very good retrieval of the true transmission in cases where no approximate methods are useful, i.e., very thin films or absorbing layers.

2) The precision of the "measured" transmission data has an effect on the accuracy of the estimation of $n(\lambda)$ but it is almost irrelevant for the estimation of $\alpha(\lambda)$. In a realistic situation using a modern spectrophotometer the transmission can be obtained with $3\frac{1}{2}$ or 4 digits. In this case, and using 100 transmission data points, the error in the estimation of $n(\lambda)$ is around 0.11 (for **Film A**). The difference between the results obtained using 100 and 50 transmission points is not meaningful.

3) In most cases the quadratic error as a function of the guessed thickness (Figs. 6 and 7) is a function with several local-nonglobal minimizers. This is an intrinsic property of this function, which might originate from a "perturbed periodic" form of the transmission. Therefore, the strategy of separating the variable d from the other variables of the

optimization problem appears to be correct, since it tends to avoid spurious convergence to those local minimizers.

4) The comparison of the present results with those previously obtained using the algorithm described in [5,6] seems to confirm that the new method is, at least, as efficient as the previous constrained optimization approach. In addition, the resulting piece of software is more portable and easier to manipulate.

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		25		50		100	
		n	α	n	α	n	α
2	$E_{ph-\min}$	0.1136	1.9021×10^{-4}	0.1063	4.3521×10^{-4}	0.1139	4.3108×10^{-4}
	$E_{ph-\max}$	0.8742	4.4408×10^{-3}	1.4727	4.9223×10^{-3}	0.4761	1.9139×10^{-3}
3	$E_{ph-\min}$	0.0442	2.7240×10^{-4}	0.1317	6.5932×10^{-4}	0.2249	1.1136×10^{-3}
	$E_{ph-\max}$	1.3505	4.4571×10^{-3}	0.2298	1.4237×10^{-3}	0.1278	1.0697×10^{-3}
4	$E_{ph-\min}$	0.0552	3.3624×10^{-4}	0.1093	4.0392×10^{-4}	0.1103	4.3715×10^{-4}
	$E_{ph-\max}$	1.3631	4.5240×10^{-3}	0.1418	1.0016×10^{-3}	0.1149	3.8185×10^{-4}
all	$E_{ph-\min}$	0.0358	3.9590×10^{-4}	0.0749	3.0955×10^{-4}	0.0184	1.6248×10^{-4}
	$E_{ph-\max}$	1.4558	4.5031×10^{-3}	0.2367	7.5514×10^{-4}	0.0117	1.9621×10^{-4}

Table I. **Film A***: Quadratic errors in the estimated refractive index and absorption coefficient with varying precision and total numbers of transmission data points.*

	25	50	100
2	121	121	121
3	119	122	124
4	119	121	121
all	118	119	119

Table II. **Film A***: Estimated thickness with varying precision and number of transmission data points.*

Film	True thickness	Estimated thickness	Quadratic error
A	118	119	6.929605×10^{-7}
B	782	782	2.203053×10^{-7}
C	147	152	6.224862×10^{-6}
D	640	639	1.365270×10^{-6}
E	624	624	2.120976×10^{-7}

Table III. True and estimated thickness and quadratic errors for all computer-generated films.

6 Figure captions

Figure 1: "True" and retrieved values of the transmission, the refractive index and the absorption coefficient of a numerically generated very thin film of thickness $d = 118\ nm$ simulating an a-Ge layer deposited on glass. Note the very good agreement found for the optical constants and the transmission.

Figure 2: "True" and retrieved values of the transmission, the refractive index and the absorption coefficient of a numerically generated film of thickness $d = 782\ nm$ simulating an a-Ge layer deposited on glass. Note the very good agreement found for the optical constants and the transmission in this absorbing film.

Figure 3: "True" and retrieved values of the transmission, the refractive index and the absorption coefficient of a numerically generated very thin film of thickness $d = 147\ nm$ simulating an a-Ge layer deposited on a crystalline silicon substrate. Note the very good agreement found for the optical constants and the transmission from an almost flat and featureless transmission spectrum corresponding to a rather narrow spectral region.

Figure 4: "True" and retrieved values of the transmission, the refractive index and the absorption coefficient of a numerically generated thin film of thickness $d = 640\ nm$ simulating an a-Ge layer deposited on a crystalline silicon substrate. Note the overall good agreement found for the optical constants and the transmission. The retrieval of the "true" index of refraction at the highest photon energies appears somewhat defective.

Figure 5: "True" and retrieved values of the transmission, the refractive index and the absorption coefficient of a numerically generated thin film of thickness $d = 624\ nm$ simulating an a-Si:H layer deposited on glass. The overall agreement for both optical constants and the transmission is very good.

Figure 6: Quadratic error of the minimization process as a function of trial thickness for **Films A** and **B**. On the left side the step is 10 nm while on the right hand side of the figure the refined step is 1 nm. Note the local-nonglobal minimizers.

Figure 7: Quadratic error of the minimization process as a function of trial thickness for **Films C** and **D**. On the left side the step is 10 nm while on the right hand side of the figure the refined step is 1 nm. Note the local-nonglobal minimizers.

Figure 8: Quadratic error of the minimization process as a function of trial thickness for **Film E**. On the left side the step is 10 nm while on the right hand side of the figure the refined step is 1 nm. Note the local-nonglobal minimizers.