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THIN FILM OPTICAL PARAMETERS DETERMINATION BY THE DYNAMICAL MODELLING AND STOCHASTIC OPTIMIZATION METHOD

We report on a new method of experimental data processing to obtain optical parameters of thin films. Dynamical modelling of the spectral reflectance can be performed interactively in a graphical environment by the genetic search in wide interval of parameter space and then refined by the genetic algorithm method, by the Nelder-Mead downhill simplex method or Marquardt-Levenberg method. Optical parameters of hydrogenated amorphous silicon (a-Si:H) thin film used in solar cell technology are determined by this new method. The spectral reflectance is a function of optical properties and the thickness of the film. Optical parameters found by our approach do not depend on the initial spectral reflectance estimation.

1. Introduction

The theoretical and experimental studies on the optical behaviour of thin films deal primarily with optical reflection, transmission and absorption properties, and their relation to the optical constants of films. The theoretical reflectance R_t can be calculated from the optical properties and thickness d of the film. Normal incidence reflectometry is often used for several reasons: good spatial resolution, high throughput and accuracy. Optical properties of any material can be described by the complex index of refraction, $N = n - ik$, where n is the refractive index and k is the extinction coefficient. Both n and k depend on the wavelength of light λ. In many thin-film applications, namely in solar cell technologies, the optical constants n and k are strongly correlated to the deposition process conditions and their broadband determination is needed to design the solar cell structures. Experimental and theoretical reflectance can be matched by fitting for the film thickness and optical properties. The optimization problem is formulated as

$$\min_{3} \left\{ \sum \left[R_o(\lambda) - R_t(n, k, \lambda, d) \right]^2 \right\} \qquad (1)$$

where R_o denotes measured reflectance values. The optimization issue is to find the system of parameters that satisfy the desired optical specifications. Numerical methods are flexible and can handle complicated problems. The numerical computations are based on merit functions that can be defined according to quantities of interest. Considering the computational cost involved, and furthermore the constraints to calculate the gradient information for this specific optical thin-film model, algorithms that can substantially reduce computation without using derivatives are preferable. In our approach the dynamical modelling and stochastic optimization approach (DMSO) is used.

2. Dynamical modelling and stochastic optimization method

The spectral reflectance (SR) of a thin quasi-ideal single homogeneous isotropic film on a thick partly absorbing substrate is given by

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

In this equation $x = \exp(-\alpha d)$ is the absorbance, $\alpha = (4\pi k_1)/\lambda$ is the absorption coefficient [1] and

$$\begin{aligned}
A &= [(1 - n_1)^2 + k_1^2][(n_1 + n_2)^2 + (k_1 + k_2)^2] \\
B &= 2[A' cos\varphi + B' sin\varphi] \\
C &= [(1 + n_1)^2 + k_1^2][(n_1 - n_2)^2 + (k_1 - k_2)^2] \\
D &= [(1 + n_1)^2 + k_1^2][(n_1 + n_2)^2 + (k_1 + k_2)^2] \\
E &= 2[C' cos\varphi + D' sin\varphi] \\
F &= [(1 - n_1)^2 + k_1^2][(n_1 - n_2)^2 + (k^1 - k_2)^2] \\
A' &= (1 - n_1^2 - k_1^2)(n_1^2 - n_2^2 + k_1^2 - k_2^2) + 4k_1(n_1 k_2 - n_2 k_1) \\
B' &= 2(1 - n_1^2 - k_1^2)(n_1 k_2 - n_2 k_1) - 2k_1(n_1^2 - n_2^2 + k_1^2 - k_2^2) \\
C' &= (1 - n_1^2 - k_1^2)(n_1^2 - n_2^2 + k_1^2 - k_2^2) - 4k_1(n_i k_2 - n_2 k_1) \\
D' &= 2(1 - n_1^2 - k_1^2)(n_1 k_2 - n_2 k_1) + 2k_1(n_1^2 - n_2^2 + k_1^2 - k_2^2) \\
\varphi &= (4\pi n_1 d)/\lambda
\end{aligned}$$

In the DMSO method, the optical parameters are determined in several steps. In the first step we prepare an initial spectral reflectance model in graphical environment by interactive dynamical modification of the reflectance model parameters. Alternatively automatic stochastic search in wide interval of parameter hyperspace can be used to create an initial reflectance model. The stochastic optimization is based on the genetic algorithm (GA). The next step uses stochastic optimization refinement of result reflectance reached in the previous modelling by the Nelder-Mead or Marquardt-Levenberg optimization:

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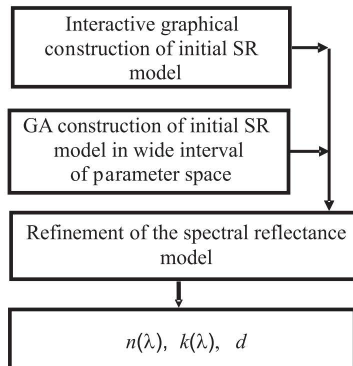

Fig. 1 Dynamical modelling method of the initial spectral reflectance model estimation

The step of dynamical modelling speeds up the calculations of the reflectance function parameters and enables constructing the theoretical reflectance model with comfort and high quality. Besides this, stochastic algorithms enable also comfortable incorporation of necessary physical constraints into the optimization method. We used genetic algorithm with binary representation of the theoretical reflectance model variables [2]. There are many different settings associated with predictions in optical parameters solutions. Most of these settings are maintained automatically using rules of the GA that create the best results. To speed up the achievement of the best solution we used visual construction of the spectral reflectance model. The values of the theoretical reflectance model parameters are dynamically modified and corresponding theoretical model is graphically displayed and compared with an experimental reflectance curve. This visual modelling step provides good estimation of the initial reflectance model and substantially reduces computation time and stochastic search of the model parameters values.

GA procedure refines the initial mathematical model by minimizing the differences between theoretical and experimental values. The initial reflectance estimation introduces a set of parameters based obviously on the properly selected dispersion model of the refractive index n and the extinction coefficient k [3,4], e.g.

$$n = n(p_1, p_2, p_3 . p_4, \lambda), \qquad k = k(p_1, p_2, p_3 . p_4, \lambda).$$

Therefore, the spectral reflectance model is represented by variables

$$X = \{p_1, p_2, p_3 . p_4, \lambda\}.$$

A set of correction values $\Delta = \{\Delta p_1, \Delta p_2, \Delta p_3 . \Delta p_4, \lambda\}$ is used in each GA iteration step for setting new trial reflectance model variables $X' = X - \Delta$. The implementation of the GA algorithm minimizes the differences (1) by stochastic exploring the correction parameters hyperspace in the interval $\langle -0.2X, 0.2X \rangle$ for each variable in the spectral reflectance function. Alternatively in case of the high quality of the initial spectral reflectance mathematical model was obtained in the first step of the DMSO method the conventional numerical optimization methods can be implemented (Marquardt-Levenberg or Nelder-Mead simplex methods):

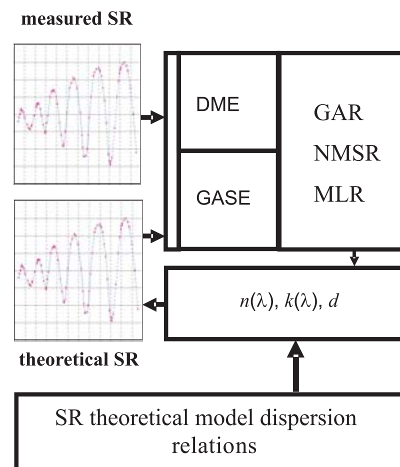

Fig. 2 DMSO method implementation
DME - Dynamical Modelling Initial SR Model Estimation
GASE - Genetic Algorithm Search SR Estimation
GAR - Genetic Algorithm method SR Refinement
NMSR - Nelder-Mead Simplex method SR Refinement
MLR - Marquardt-Levenberg method SR Refinement

3. Experimental results

The described method was used to extract the refractive indices of a series of undoped a-Si:H thin films with approximately the same thickness ~ 400 nm (Table 1). The samples were deposited at the Delft University of Technology, the Netherlands, on Corning 1737 glass substrates by PECVD industrial deposition system [5,6] from hydrogen (H_2) to silane (SiH_4) plasma under varied H_2/SiH_4 gas flows (the dilution D).

Samples under study Table 1

dilution D	thickness [nm]
0	390
10	394
20	385
30	388
40	402
50	397

Optical properties of the dilution series are obviously affected by the changing structure [6]. The refractive indices for all samples under study were determined from the experimental spectral reflectance determined by the DMSO method (Fig. 3). At all wavelengths, we observe the decreases of the refractive index with increasing dilution For the sample prepared at $D = 40$, approximately the same spectral refractive indices were revealed as for the

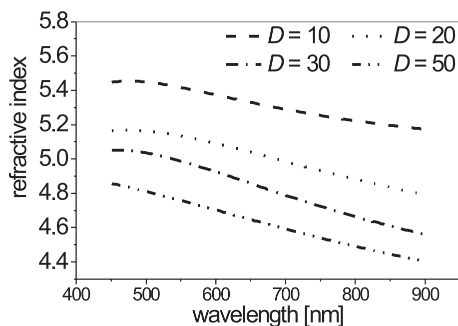

Fig. 3 Refractive indices of the a-Si:H thin film samples.

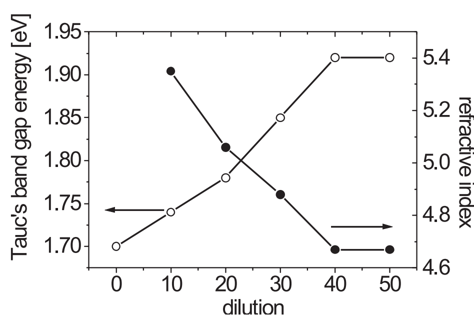

Fig. 4 Refractive index (at 633 nm) and the Tauc's optical band.

sample at $D = 50$, therefore this plot was not incorporated into the Fig. 3.

Fig. 4 illustrates the dilution-dependent refractive index at 633 nm and Tauc's optical band gap determined from the experimental spectral transmittance in the vicinity of the absorption edge [6]. The data for Tauc's optical band gap as one member of the set of dispersion model parameters acquired from the course of DMSO method are in good agreement with the values plotted in Fig. 4.

Thus, the DMSO method yields quickly and reliably information useful for all applications of the samples under study where optical properties matter. The observed decrease of refractive index may be probably due to the voids – sites with missing radicals of $SiH_4 - H_2$ decomposition. The band gap appears to increase with increasing dilution. The increase of E_g at the amorphous-crystalline boundary is due to the appearance of indirect gap electronic structure of the crystalline silicon.

4. Conclusions

The DMSO method was introduced for the thin film optical parameter determination. Experimental spectral reflectance data of a series of a-Si:H thin films were analysed. DMSO method succeeded in theoretical reflectance model refinement very good. It does not converge to a local optimum of the refined model. The combination of dynamical modelling and stochastic optimization of the initial mathematical model compared with the experimental data speeds up calculations and enables checking for meaningful optical parameters.

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