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Hybrid GA-gradient method for thin films ellipsometric data evaluation

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ABSTRACT

A global-search method which applies the concept of genetic algorithm (GA) with gradient-based optimizer is proposed for the problem of experimental data analysis from spectroscopic ellipsometry on thin films. The method is applied to evaluate the data obtained for samples with different structure complexity, starting with transparent monolayers (SiO_2, HfO_2) on a substrate, through absorbing film (diamond-like carbon) and multilayer structures. We demonstrate that by using this method we are able to find material parameters even for limited a priori knowledge about the sample properties, where classical methods fail.

1. Introduction

Spectroscopic ellipsometry (SE) is nowadays a well-established optical technique, routinely applied to characterize material properties at both research and industrial scale. Taking advantage of light polarization state change upon interaction with the sample under studies [1,2], SE allows non-destructive and contactless determination of the material optical parameters – real and imaginary parts of the complex refractive index from a single measurement, and films thicknesses with sub-nanometers sensitivity.

In ellipsometry, the measured light polarization change upon interaction with the sample is described by two parameters, (Ψ, Δ), which express the amplitude ratio change and phase shift, respectively, between *p*- and *s*-components of the polarized light. Once the (Ψ, Δ) parameters are measured, the task is to determine all the sample parameters of interest, i.e. optical and geometrical properties. Due to the lack of inverse equations, this cannot be done directly from the experimental spectra. Except the fundamental case of light interaction at a single smooth interface (bulk sample with perfect surface), the so-called inverse problem need to be solved in ellipsometry, in which the appropriate analytical model of the studied sample is assumed and the calculated optical response of the model is compared to the experimental data. By adjusting the model, the difference (error) between experimental and modeled data is minimizing and the desirable material parameters can be obtained.

The described process can be considered as a classical global optimization task. To solve the problem, a linear regression method with use of a gradient-based Levenberg–Marquardt algorithm (LMA) [3] is routinely applied. However, as for all gradient methods, the efficiency of LMA is strongly dependent on the choice of good initial points for the search process. The necessity to deliver initial values of the model parameters close to the true sample parameters, which often means the use of complementary experimental methods, clearly limits the usefulness to the ellipsometry method for the studies of complex and/or new materials [4–7]. Tedious search for the relevant starting points is a true nightmare of each ellipsometry experimentalists.

It is well known that if the error surface is nonquadratic, non-monotonously decreasing, after several iterations the gradient methods usually slip into one of the local minima, instead of finding the global one. Because of the non-linearity and complexity of the model equations, the error surface in ellipsometry is mostly rough [8]. Therefore, a more complex global-search methods would be desirable for the purpose of ellipsometry data evaluation. Among different global-search methods, metaheuristic optimization algorithms have already attracted much attention in different fields of science and technology [9]. The biggest advantage of that type of search algorithms is that they do not make any assumptions about underlying fitness landscape, avoiding the disadvantages of iterative improvement and, in particular, multiple descents by allowing the local search to escape from local optima [10]. At the same time, as not being just a simple random methods but rather a complex learning processes, this type of algorithms ensure much more reliability comparing to pure heuristics.

Therefore, it is not surprising that there were already some attempts to use metaheuristic optimization methods also in ellipsometry. In

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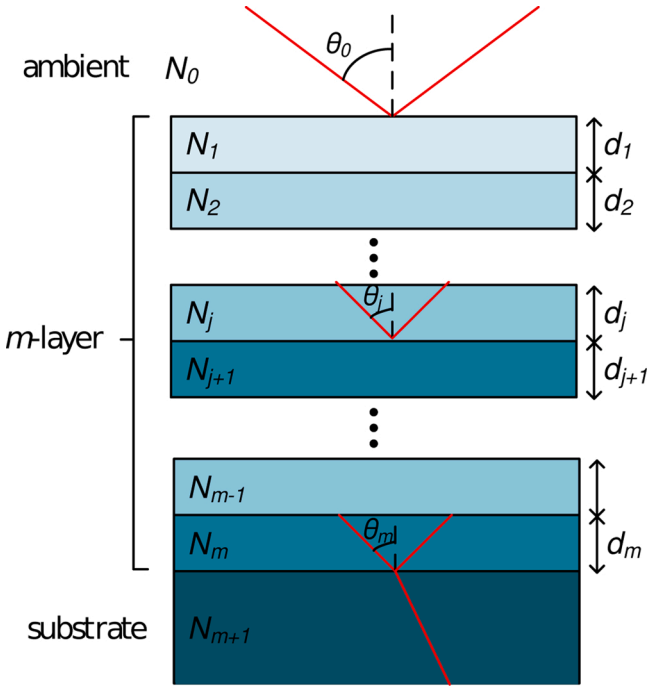

Fig. 1. A schematic representation of a m-layer stratified structure sandwiched between two semi-infinite ambient (0) and substrate ($m+1$) media. Symbols $d_{\#}$, $N_{\#}$ and $\theta_{\#}$ denote thicknesses, complex refractive indices, and angles of refraction respectively, for each media.

particular, in early works of Polgar et al. [8,11,12], the authors applied some random-search methods together with genetic and simulated annealing algorithms to solve ellipsometric problem for relatively complex optical models. However, the main drawback which the authors had that time was a high convergence time of the algorithms and in some cases limited reliability. Later attempts deal with rather small searching space – relatively small number of fitting parameters (low complexity of the model) or/and narrow searching ranges [13,14], what calls into question the necessity of using global-search methods in all these cases. Other works [15,16] show promising results for the application of metaheuristics in ellipsometry but they are only limited to the studies of particular materials rather than the comprehensive discussion about the usefulness of that kind of methods in the context of ellipsometry. As a result, the question about the capabilities and limitations of metaheuristic optimization methods for ellipsometry is still open.

In this work, we apply a method which combines the concepts of genetic algorithm (GA) and gradient-based search methods for ellipsometry data analysis. The proposed hybrid genetic-gradient algorithm (further denoted as HGGA) is tested on representative ellipsometry problems, starting with a single transparent film at known substrate up to the more complex issues of ellipsometry on absorbing films and multilayer structures. The main idea of HGGA is that, as it uses a global-search method, the algorithm shall be able to determine material parameters without need to deliver any initial values (no a priori knowledge about sample properties) in the process of ellipsometry data evaluation.

2. The method

In the problem of ellipsometry data analysis, the theoretical model of the studied sample – optical model, is built as a system which consists of a plan-parallel layers. Each layer is described by its optical parameters and thickness. Afterwards, the theoretical values of (Ψ, Δ) parameters are calculated from the model over the analyzed spectral range.

As we restricted our considerations to isotropic materials, the optical response of the m-layer stacked model (see Fig. 1) can be calculated using a 2×2 transfer matrix method [2,17,18]. In this method, the sample total transfer matrix $\mathbf{S}_{s|p}$ for s- or p-polarization is defined by:

$$\mathrm{S}_{s|p} = \mathrm{I}^{01}_{s|p}\cdot\mathrm{L}^{1}_{s|p}\cdot\mathrm{I}^{12}_{s|p}\cdot\mathrm{L}^{2}_{s|p}\cdot\ldots\cdot\mathrm{I}^{j-1,j}_{s|p}\cdot\mathrm{L}^{j}_{s|p}\cdot\mathrm{I}^{j,j+1}_{s|p}\cdot\ldots\cdot\mathrm{I}^{m-1,m}_{s|p}\cdot\mathrm{L}^{m}_{s|p}\cdot\mathrm{I}^{m,m+1}_{s|p} \tag{1}$$

where $\mathrm{I}^{j,j+1}_{s|p}$ is a transfer matrix between j-th and j+1 interface for s- or p-polarized light:

$$\mathrm{I}^{j,j+1}_{s|p} = \frac{1}{t^{j,j+1}_{s|p}}\begin{bmatrix} 1 & r^{j,j+1}_{s|p} \\ r^{j,j+1}_{s|p} & 1 \end{bmatrix} \tag{2}$$

and $L^{j}_{s|p}$ is a transfer matrix of the j-th layer for s- or p-polarization:

$$\mathrm{L}^{j}_{\mathrm{sp}} = \begin{bmatrix} exp(-i\beta_j) & 0 \\ 0 & exp(i\beta_j) \end{bmatrix} \tag{3}$$

The term β_j is given by the formula:

$$\beta_j = \frac{2\pi d_j}{\lambda} N_j cos(\theta_j) \tag{4}$$

where d_j, N_j, θ_j are thickness, complex refractive index and angle of refraction, respectively, of the j-th layer, and λ is the wavelength of light. Symbol $r^{(j,j+1)}_{(s|p)}$ $\left(t^{(j,j+1)}_{(s|p)}\right)$ denotes the amplitude reflection (transmission) coefficient at $(j, j+1)$ interface given by Fresnel equations:

$$\begin{aligned} r_p^{j,j+1} &= \frac{N_{j+1}cos(\theta_j) - N_j cos(\theta_{j+1})}{N_{j+1}cos(\theta_j) + N_j cos(\theta_{j+1})} \quad r_s^{j,j+1} = \frac{N_j cos(\theta_j) - N_{j+1}cos(\theta_{j+1})}{N_j cos(\theta_j) + N_{j+1}cos(\theta_{j+1})} \\ t_p^{j,j+1} &= \frac{2N_j cos(\theta_j)}{N_{j+1}cos(\theta_j) + N_j cos(\theta_{j+1})} \quad t_s^{j,j+1} = \frac{2N_j cos(\theta_j)}{N_j cos(\theta_j) + N_{j+1}cos(\theta_{j+1})} \end{aligned} \tag{5}$$

Based on Eqs. (1)–(7) and taking into account that the total reflection coefficient $r^{012\ldots m}_{s|p}$ of any m-layer optical model is equal to the sample transfer matrix elements $\mathrm{S}^{21}_{s|p}/\mathrm{S}^{11}_{s|p}$, analytical expression for $r^{012\ldots m}_{s|p}$ can be derived for p- and s-polarization, which in turns relates to the ellipsometric (Ψ, Δ) parameters by the known formula:

$$tan(\Psi)exp(i\Delta) = \frac{r_p^{012\ldots m}}{r_s^{012\ldots m}} = \frac{S_p^{21}}{S_p^{11}} \times \frac{S_s^{11}}{S_s^{21}} \tag{6}$$

Having the calculated optical model response, the task is to find the set of the unknown model parameters (layer's thicknesses and optical constants) for which the model values of (Ψ_{mod}, Δ_{mod}) parameters fit the corresponding experimental ones (Ψ_{exp}, Δ_{exp}) over the analyzed spectral range. To evaluate the quality of fit we use a standard fitting error function for ellipsometry, being the modified root mean squared error (*RMSE*) of the following form:

$$\mathrm{RMSE} = \sqrt{\frac{1}{2M}\sum_{j=1}^{M}\left(\frac{\Psi_{\exp}(E_j) - \Psi_{\mathrm{mod}}(E_j)}{\delta\Psi(E_j)}\right)^2 + \left(\frac{\Delta_{\exp}(E_j) - \Delta_{\mathrm{mod}}(E_j)}{\delta\Delta(E_j)}\right)^2} \cdot 1000 \tag{7}$$

where ($\delta\Psi$, $\delta\Delta$) show measurement errors in (Ψ, Δ) [1] and subscripts *exp* and *mod* denote experimental and modeled, respectively, parameters at jth spectral point (jth photon energy E_j), and M is the number of spectral points. Thus the aim of the optimization process is to minimize the objective function given by Eq. (7).

In the method proposed here, we developed an algorithm which combines the concept of GA with a gradient-based search method. Based on the GA principle [19,20], the search process starts with a number of random values for the model parameters, where each set of the individual parameter values is called a chromosome. All chromosomes (population) are evaluated using a fitness function (Eq. (7)). According to a natural selection process that mimics biological evolution, those

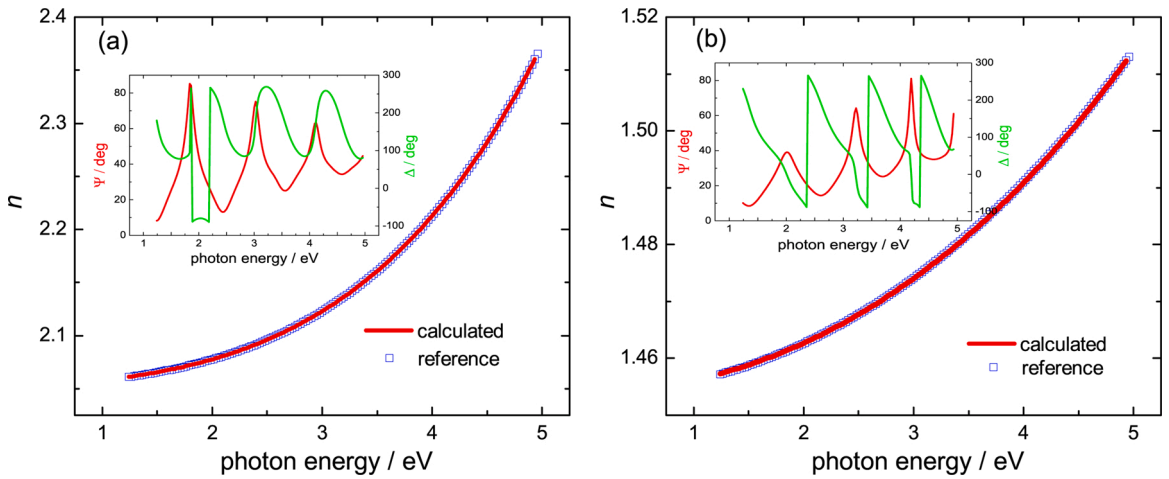

Fig. 2. Reference [23,24] (symbols) and calculated by HGGA (solids) refractive index for (a) HfO_2 and (b) SiO_2 thin films. Insets show corresponding simulated ellipsometric spectra for 250 nm and 450 nm thick HfO_2 and SiO_2, respectively, on Si.

chromosomes with the lowest *RMSE* (elite individuals) are kept in the course of generating a new population. The less adaptive ones are modified by two GA operators: crossover and mutation. The first one combines two selected chromosomes (parents) at random from the current population to form new individuals (children) for the next generation. Mutation rules in turn apply random changes to individual parents to form children. The whole process is then repeated until either the assumed maximum number of generation has been reached or the changes in *RMSE* are little.

It is known that, as GA can relatively quickly reach the neighborhood of the global search minimum, it can require many objective function evaluations to achieve convergence at the exact minimum [21]. First trials showed also that if the search space is flat around the solution, GA may return results which are far away from the true values of the actual parameters. To avoid the problem described and to speed up the solution process, we applied the scheme in which the GA is first run for a relatively small number of generations. Then the parameter values obtained from GA solutions are used as a starting point for the gradient-based local optimizer. This kind of hybrid genetic-gradient algorithm (HGGA) allows to compromise between two conflicting objectives which are exploiting the best solutions found so far and at the same time exploring the search space for promising solutions [22].

The search algorithm together with the respective model equations was implemented in Matlab®. We used a standard GA implementation from the Global Optimization Toolbox. The controlled parameters of the GA were: the number of generations, size of the population (number of candidate solution at each iteration) and a crossover fraction, where the last one defines the amount of individuals other than elite which exhibit crossover (i.e. crossover fraction 0.6 means that 60% of the children are crossover children when the rest are mutation children). The fraction of population kept as elite was set to 5%. Roulette wheel was chosen as a selection function [19] and default Matlab *Scaterred* function for crossover. Moreover, the Matlab *Gaussian* was taken as a mutation function in which the way how the mutation range is changing over the generations can be controlled by the parameter called *Shrink*, which controls how the standard deviation σ shrinks at the kth generation according to formula:

$$\sigma_k = \sigma_{k-1}\left(1 - \text{Shrink}\frac{k}{\text{Generations}}\right) \tag{8}$$

For the final local optimization we employed the *fminunc* function from the Optimization Toolbox which uses known quasi-newton Broyden–-Fletcher–Goldfarb–Shanno (BFGS) algorithm. All the calculations has been done using standard PC equipped with 2.4 GHz i5-6200U CPU and 8 GB RAM.

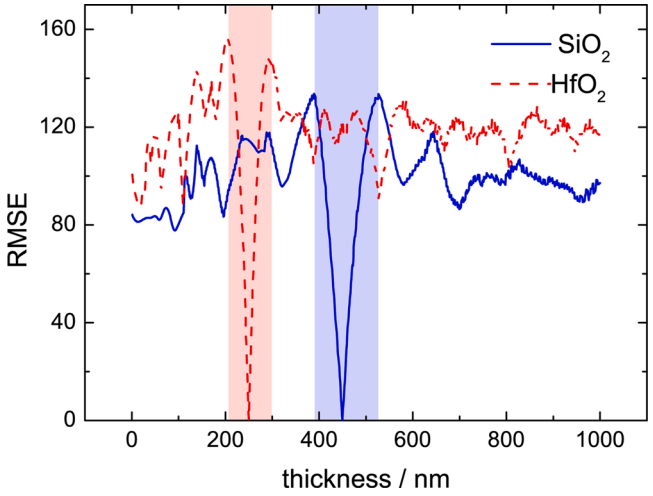

Fig. 3. Search function for 450 nm thick SiO_2 (solid) and 250 nm thick HfO_2 (dash) films on Si for the case when the thickness of the film is the only search parameter. Shaded regions indicate that range of initially assumed parameter values which allows gradient-based methods to find the minimum.

3. Validation of the method

In this section, the effectiveness of the proposed HGGA method is demonstrated on typical problems in ellipsometry data analysis of thin films. The capabilities of the method to determine the optical constants and thickness of single transparent/absorbing film and multilayer structures – without necessity to deliver initial values for the search process, is tested for several simulated and real samples.

3.1. Transparent film

To demonstrate the capabilities of the proposed search algorithm in ellipsometry on transparent films we have chosen two reference systems: (1) thermally grown silicon dioxide SiO_2 and (2) hafnium dioxide HfO_2, both films deposited on silicon substrate Si. The reference optical constants [23,24] of the films and calculated optical response of the simulated samples are shown in Fig. 2. According to Cormier et al. [25], presence of Gaussian noise with standard deviation of 0.01° and 0.02°, which is the typical accuracy for standard rotating analyzer ellipsometer [26], was added to the Ψ and Δ spectra, respectively, to reproduce experimental conditions.

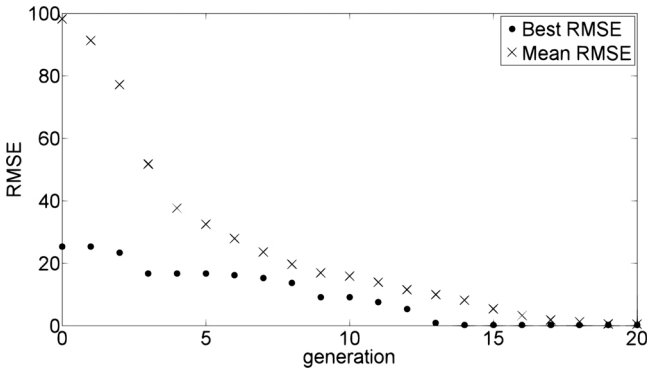

Fig. 4. Evolution of the *RMSE* value as the generation increases for SiO_2/Si when the thickness of the layer is the only search parameter.

We started our considerations with the simplest situation in the ellipsometry data analysis of thin films in which the refractive index *n* of each material is known. In this case, the thickness of the layer *d* is the only parameter which need to be determined. In Fig. 3 the change of *RMSE* with the variation of the thickness – an objective function, is demonstrated for both studied reference systems. The plot is representative for all that type of ellipsometric problems. As it can be easily seen from Fig. 3, the objective functions in this case possess a distinct minimum with the lowest *RMSE* value – a global minimum, which points to the searched layer thickness. It is obvious, that the classical gradient-based search algorithm is able to solve this problem only if one supplies initial thickness values from the narrow range in the neighborhood of the global minima (see shaded regions in Fig. 3). Otherwise, the gradient algorithm gets stuck at one of the local minima with clearly higher value of *RMSE*.

In Fig. 4, an example result of the application of HGGA to the described problem is shown. The algorithm converges to the solution after about 15 generations with time not more than 0.6 s. The reproducibility of the solution is ensured for the minimum population size of 70. The obtained results are 449.94 and 249.97 for nominal 450 nm SiO_2 and 250 nm HfO_2, respectively. Small discrepancies between the calculated and nominal values are due to simulated noise in the (Ψ, Δ) spectra. It is obvious that this particular case when the thickness of transparent layer is the only fitting parameter, good starting point for LMA or other gradient methods can be found relatively fast by careful analysis of the number of oscillations seen in experimental data, by plotting the objective function, or by just simple guessing. Nevertheless, the proposed global-search method allows to avoid tedious search for the correct initial value of the thickness and at the same time to fully automatize the analysis process.

At next step, the HGGA method is tested for the situation where not only the thickness *d* of the transparent film is unknown but also its refractive index *n*. It is important to emphasize that this problem cannot be solved unambiguously when the experimental data are analyzed for each experimental point independently (so called point by point fit). Due to the strong correlation between thickness and refractive index there exists periodically distributed multiple global minima in the objective function. Therefore, if the parameter search ranges are relatively large, parameterization of the refractive index is needed to ensure a unique fit.

The dispersion of refractive index of common transparent materials can be described by the well-known Cauchy formula:

$$n(\lambda) = A + \frac{B}{\lambda^2} + \frac{C}{\lambda^4} \tag{9}$$

where *A* relates to the amplitude ratio, and *B* and *C* provide the curvature of the refractive index towards short wavelengths λ. As in this case there are four search parameters (3× Cauchy parameters and film thickness), the solution need to be found in the 4-dimensional search space. To illustrate the problem, part of the search space for 450 nm SiO_2/Si has been plotted in Fig. 5 as an example. It is clearly seen in Fig. 5 that the search space is rough with multiple local minima (yellowish parts). At the same time a global minima region (dark blue) can be distinguished for this class of ellipsometric problem, thus, the values of the search parameters can be found unambiguously. Furthermore, the picture clearly shows that a global-search method is desirable when the knowledge about the studied sample is limited, i.e. the search parameter space is large.

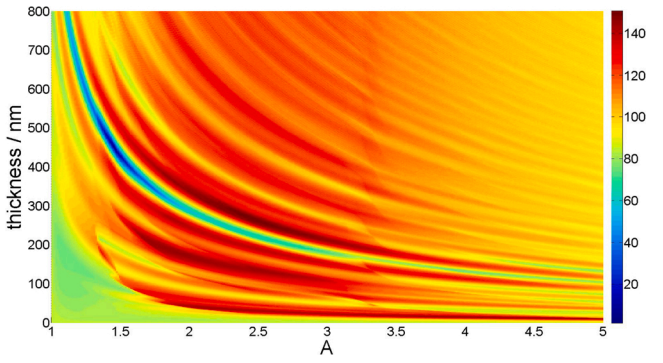

Fig. 5. An example part of the search space (dependency of *RMSE* on the Cauchy parameter *A* and the layer thickness) for reference 450 nm thick thermally grown SiO_2 on Si.

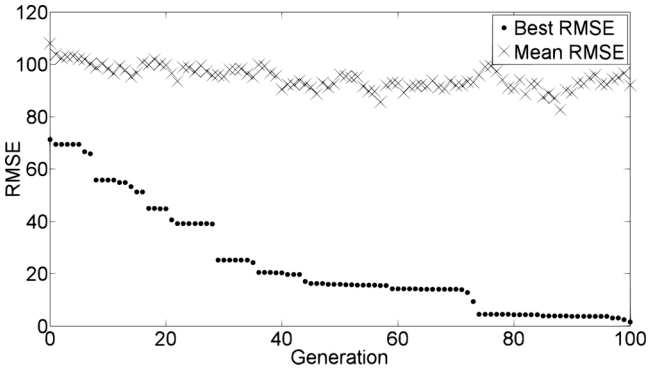

Fig. 6. Evolution of *RMSE* value as the generation increases for SiO_2/Si – thickness and Cauchy model parameters are the unknown. Good starting values for the gradient algorithm are found after about 80 generations. High mean RMSE value over the whole generations is due to the mutation *Shrink* parameter set to 0.5.

Table 1
The determined values of the Cauchy parameters *A*, *B*, *C* and film thickness for the studied reference transparent films.

Sample	*A*	*B*	*C*	*d* [nm]	*RMSE*
250 nm HfO_2/Si	2.052	0.0076	0.0007	249.97	1.07
450 nm SiO_2/Si	1.454	00033	0	450.03	1.20
1000 nm SiO_2/Si	1.452	0.0029	0	1007.9	1.19
25 nm SiO_2/Si	1.503	0.02	0	22.53	1.11

An example convergence curve for 100 generations is shown in Fig. 6. The curve is representative for both reference samples at all algorithm runs. The genetic algorithm needs not more than 80 generations with the population size of 200 to find good initial points for the gradient algorithm, which subsequently localizes the global minimum with the objective function value of 1.2 and 1.07 for SiO_2 and HfO_2, respectively (total execution time about 4 s). The determined refractive indices for both films are in agreement with the corresponding reference

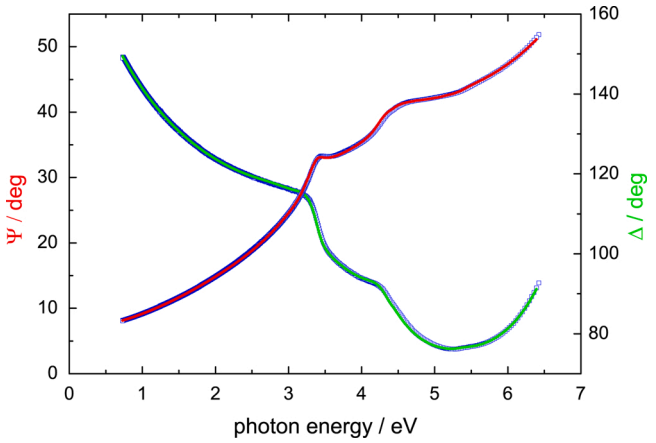

Fig. 7. Experimental (symbols) and calculated (solid) ellipsometric spectra for Si with thermally grown SiO_2 of nominal thickness 25 nm.

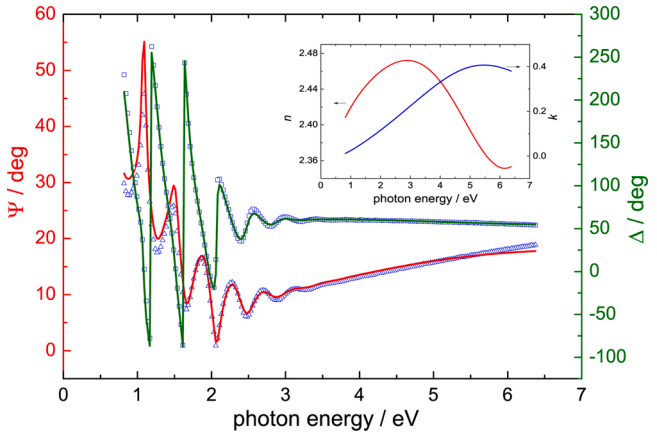

Fig. 8. Experimental (dots) and modeled (solids) ellipsometric $\Psi(E)$, $\Delta(E)$ spectra for 489 nm thick DLC film on SW7M steel. Inset shows the determined refractive index n and extinction coefficient k.

data (see Fig. 2). All the obtained model parameters are collected in Table 1. To avoid convergence to a local minimum at early generation, the algorithm keeps high mutation range by setting the shrink parameter to 0.5. This, together with the empirically determined best crossover fraction value of 0.6, allows a good reproducibility of the method for the discussed ellipsometric problem.

Finally, the proposed method is tested at real experimental conditions. For this purpose we used two Si wafers with thermally grown SiO_2 delivered by Woollam Co., with nominal thickness of SiO_2 of around 1000nm and 25 nm. Ellipsometric spectra were measured by a Woollam RC2 dual-compensator ellipsometer in the spectral range 0.73–6.4 eV with spectral stepwidth of 0.04 eV and the light angle of incidence 70°. The HGGA method with the same algorithm parameters as in the example above was applied to find the Cauchy dispersion model parameters and thicknesses of both films. No assumptions about the expected model parameters values are made thus the films are treated as completely unknown.

An example for experimental and calculated (Ψ, Δ) spectra for SiO_2 with the nominal thickness of 25 nm are shown in Fig. 7. The obtained model values are collected in Table 1. The calculated (Ψ, Δ) spectra fit very well the corresponding experimental ones. The *RMSE* evolution curve with generation has similar character to that shown in Fig. 6. Hereby, the effectiveness of the proposed method for ellipsometry data analysis of transparent films was proven.

Here, it shall be emphasized that the method does not make any assumptions about the expected model parameters values. This means not only that the algorithm do not need any initial values of the parameters to start the search process, but also no assumption about the expected range of values of the fit parameters. In the case of transparent films data analysis the search space possess rather distinct global minimum (see Figs. 3 and5). Thus, randomly generated unphysical or unreliable values of the model parameters are rejected at the initial steps of the search process as those with wrong fitness function values (extremely high *RMSE*). As a result, the method can be successfully applied to evaluate ellipsometric data in the situation of very limited knowledge about the physical properties of the studied material which is not the case of the classical gradient-based methods.

3.2. Absorbing film

As there are certainly many situations where the film can be treated transparent, in many cases transparency of the film cannot be assumed a priori. Even in the presence of periodic oscillations in the ellipsometric data, being the result of light interference reflected from the top and bottom of the film, in many cases some weak absorption is needed to be taken into account during the evaluation of the ellipsometric data. Likewise, if the film is relatively thin (usually less than about 100 nm for most absorbing materials), light will reach the film/substrate interface also when the film absorption coefficient is relatively high.

To test our method on the absorbing film, we have measured ellipsometric spectra for diamond-like carbon (DLC) coating deposited on SW7M stainless steel. The film was deposited by a combined DC and high power impulse vacuum-arc process. Details on the sample preparation can be found elsewhere [27]. DLC is a well-known material used as a protective or antireflective coating in tribological, optical and biomedical applications and others. Its optical properties strongly dependent on the deposition method and process conditions [28], substrate used [29], content of sp^2 and sp^3 bonded carbon [30] etc. Thus, there are no good reference data for DLC optical constants available.

The experimental $\Psi(E)$, $\Delta(E)$ spectra are shown in figure 8 (symbols). Oscillations in low-energy parts of the spectra clearly indicate interferences of the light reflected from the top and bottom of the film. However, as the amplitude of interference oscillations is decreasing towards higher energies, absorption of the film need to be taken into account during data evaluation. Therefore, optical spectra of the film were parameterized by the Tauc–Lorentz (T–L) dispersion formula [31] relevant to amorphous materials, where imaginary part of the dielectric function is given by:

$$\varepsilon_2(E) = \begin{cases} \dfrac{AE_0C\left(E-E_g\right)^2}{\left(E^2-E_0^2\right)^2+C^2E^2}\cdot\dfrac{1}{E} & E > E_g \\ 0 & E \le E_g \end{cases} \tag{10}$$

where A and C are amplitude and broadening parameters (fitting parameters), E_g is the optical band gap, and the real part of the dielectric function can be obtained by Kramers–Kronig transformation (see erratum to [31] for analytical solution). Optical constants of the

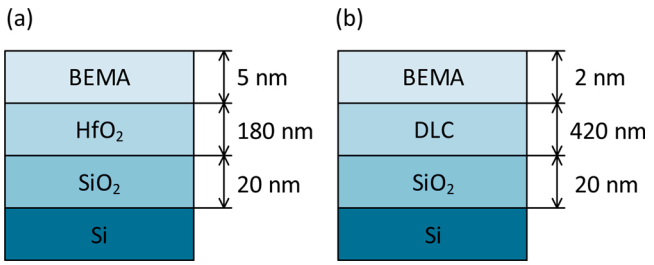

Fig. 9. Reference multilayer structures: (a) two transparent layers SiO_2 and HfO_2 sandwiched between Si substrate and surface roughness layer (BEMA), and (b) HfO_2 layer is replaced by absorbing DLC.

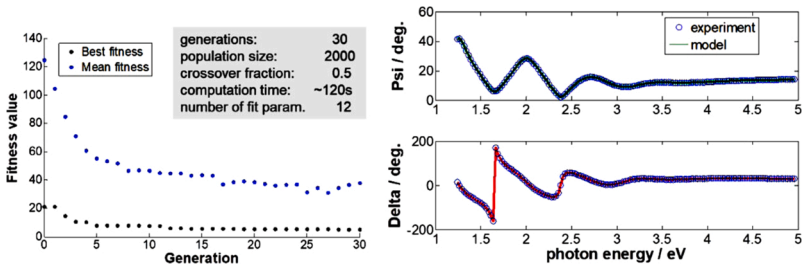

Fig. 10. An example HGGA convergence curve for complex multilayer structure (BEMA/DLC/SiO_2/Si) – left side, and the corresponding experimental (dots) and modeled (solids) ellipsometric $\Psi(E)$, $\Delta(E)$ spectra – right.

substrate were determined from the numerical inversion of the ellipsometric data obtained on bare SW7M stainless steel. Additionally, presence of surface roughness was modeled as a top layer with optical constants described by standard Bruggemann effective medium theory (BEMA) assuming 50% of void [32].

The HGGA method was applied to find 8 in total model parameters: 6× T–L formula parameters and thickness of the DLC film d_{DLC} and roughness layer d_r. Once again, no assumption about the expected search parameter values was made. The convergence curve for each generation has similar character to that shown in Fig. 6. The algorithm is able to find the best match model parameters values ($RMSE = 3.17$) after around 100 generations with population size of 500 and in time of about 5 s. for one algorithm run. The obtained DLC optical constants and the corresponding model ellipsometric spectra are shown in Fig. 8. The determined thicknesses of the DLC layer is 489 nm.

For this particular problem, for certain algorithm runs the method returned results with visibly higher value of *RMSE*, usually in the range between 10 and 20. These results indicate for the existence of some local minima in the search space with objective function values close to the found global one. This shows another interesting feature of the method. As it uses metaheuristics, the way how the search space is explored is varying from one to another algorithm run. Therefore, by multiple re-run of the algorithm other candidate solutions can be found in the case of complex problems with non-unique solution. At the end, the role of human-expert would be to judge which result is the most reliable.

3.3. Multilayer structures

To further check the capabilities of the method, we have tested it for a relatively complex problem where multi-layer samples with large number of the unknown parameters are evaluated. For this purpose, ellipsometric data are simulated for two model structures as shown in Fig. 9. For the first structure, presence of two transparent layers on Si: 180 and 20 nm thick HfO_2 and SiO_2, respectively, together with 5 nm BEMA layer describing the surface roughness, is assumed. Accordingly, for the second structure, one transparent layer is replaced by absorbing 420 nm thick DLC layer.

To find the optical constants of each modeled layer, the dispersion of the complex refractive index is described, as above, by Cauchy and T-L formula for transparent and absorbing layers, respectively. As a result, the task is to find 9 in total (2×3 Cauchy parameters + thicknesses of each layer) parameters for the structure 1 and 12 parameters (6× T–L + 2× Cauchy + thicknesses) for the structure 2. As in this case the search space is much more complex than for single layer samples, the search parameters were bound in the reasonable ranges to prevent the exploration parts of the search space from assuming unreliable and non-physical parameter values (negative thicknesses, enormous values etc.).

An example results of the application of HGGA to structure 2 are shown in Fig. 10. The determined thicknesses of the layers are: 5, 180.02, 19.99 nm for roughness and two transparent layers, respectively, for the structure 1; and 1.96, 19.36, 418.74 nm for roughness, absorbing and transparent layer, respectively, for the structure 2. It was found that setting the relatively large population size of about 2000 clearly increases the chance to find the values of the unknown model parameters. Once again, due to the complex multi-dimensional search space and broad search ranges, even then several runs of the algorithm is necessary to find the best-matched results (computation time for one algorithm run is about 50s and 120 s for the structure 1 and 2, respectively). However, finally the algorithm returns results with visibly lower value of the *RMSE* than for the other runs. Therefore, this test proves that the proposed method can be successfully applied even for relatively complex ellipsometric problems, which can be solved by classical gradient-based method only if good initial parameter values are delivered.

4. Conclusion

In this work, a hybrid genetic-gradient algorithm (HGGA) was developed for the purpose of experimental data evaluation from spectroscopic ellipsometry on thin films. This method uses the genetic algorithm concept to explore the objective space; hence do not make any assumptions about the underlying fitness landscape, avoiding the disadvantages of iterative improvement and, in particular, multiple descents by allowing the local search to escape from local optima. Afterward, the gradient-based algorithm is employed to explore the promising part of the objective space to find the global *RMSE* minimum.

The method was successfully applied to representative ellipsometric problems of different complexity. The algorithm allows rapid and reliable determination of the complex refractive index and thickness of the studied transparent and absorbing layers. The main benefit is that, in contrary to gradient-based search algorithms – like the ellipsometry traditional Lavenberg–Marquardt algorithm, the proposed method do not need any starting values to initialize the search process. As a result, it can be successfully applied in situation of limited preliminary knowledge about the properties of the studied sample. This method has also another interesting feature. As being global-search and non-deterministic, HGGA has an ability to return multiple candidate solutions in case of complex search space with non-unique extrema, which can be further evaluated by human-expert.

In summary, the obtained results show metaheuristic optimization methods as promising alternative for derivative-based search methods for the purpose of spectroscopic ellipsometry data evaluation.

Conflict of interest

None declared.

Declaration of Competing Interest

The authors report no declarations of interest.

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