Research Article

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Combination of a global-search method with model selection criteria for the ellipsometric data evaluation of DLC coatings

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Abstract: A method for the evaluation of experimental data from spectroscopic ellipsometry is proposed which combines the global-search optimization algorithm with statistical model selection criteria. The hybrid genetic-gradient search algorithm (HGGA) is applied to find the optical parameters and thickness of a diamond-like carbon (DLC) coating deposited on SW7M stainless steel. Akaike and Bayesian information criteria are used to evaluate the different dielectric function models. The method is able to find optical model parameters even in case of a limited initial knowledge about the material optical constants. At the same time, the optimal dielectric function model for the description of the material optical properties can be selected unambiguously from the set of candidate models.

Keywords: data evaluation; genetic algorithm; information criteria; spectroscopic ellipsometry.

1 Introduction

The indirect character of Spectroscopic Ellipsometry (SE) results in the necessity of non-trivial, in general numerical analysis of the experimental data in order to obtain desired material parameters. This process aims to provide an appropriate analytical model of the studied sample and subsequently to fit the modeled and experimental data [1]. For this purpose, classical gradient-based optimization algorithms, like the Levenberg-Marquardt, are commonly used, where the minimum difference (error) between experimental and modeled data is the only optimization criteria [1].

Due to the complexity and nonlinearity of the equations used for the modeling of the ellipsometric experiment, fitness function commonly possesses either multiple global minima at which search parameters obtain different values or local minima causing the gradient-based search algorithm to get stuck. Recently, we proposed the application of an evolutionary optimization algorithm in SE [2]. In work [2], a global-search hybrid gradient-evolutionary algorithm, which applies the concept of genetic algorithm (GA) for the fully exploration of the search space, has been applied to the problem of experimental data analysis from SE on thin films. The method allows for the determination of material parameters even for limited *a priori* knowledge about studied sample parameters, where classical methods fail [2].

Nevertheless, the effectiveness of any search algorithm is strongly conditioned by the existence of a unique global extreme in the search space of parameters variations. In many cases, fulfilling of this criterion is only possible by the parameterization of ellipsometric spectra by one of the optical dispersion models. At this point, the open question is, which from the variety of parametric dispersion models would be the best choice for the given problem? In works [3–5], Likhachev suggested the application of two well-established statistical methods for model selection, namely, Akaike (*AIC*) and Bayesian (*BIC*) Information Criteria (IC), to compare different dispersion models with various complexities. This approach allows to determine objectively the "best" one from a set of candidate models, avoiding under- or overfitting of experimental data.

The combination of the mentioned criteria with the metaheuristic global-search optimization algorithm may result in solving of two main problems of ellipsometry data analysis, namely: (1) selecting the appropriate dispersion model, and (2) delivering "good" starting points for search algorithm. Following this idea, in this work we combined

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the *AIC* and *BIC* information criteria with a hybrid gradient-evolutionary algorithm (HGGA) [2]. The proposed method is applied to the data evaluation of ellipsometric spectra from diamond-like carbon (DLC) coatings and its effectiveness is discussed.

2 Method details

The problem of spectroscopic ellipsometry data analysis can be considered as a consecutive order three-phase process [6]. First, the theoretical model of the studied sample – optical model, is built as a system of plan-parallel layers in general, where each layer is described by its optical parameters and thicknesses. Then, the optical response of the model can be calculated using a 2×2 transfer matrix method [1, 7, 8]. Finally, the task is to fit the theoretical ellipsometry spectra, typically $\Psi(E)$ and $\Delta(E)$, where E is the photon energy, to the experimental ones by varying the model parameters. The whole process can be considered as an optimization task in which the goal is to minimize objective function, being the optimization criteria formulated as a modified root mean squared error (*RMSE*) of the following form:

$$RMSE = \sqrt{\frac{1}{2n}\sum_{j=1}^{n}\left(\frac{\Psi_{\exp}(E_j)-\Psi_{mod}(E_j)}{\delta\Psi(E_j)}\right)^2+\left(\frac{\Delta_{\exp}(E_j)-\Delta_{mod}(E_j)}{\delta\Delta(E_j)}\right)^2} \quad (1)$$

where ($\delta\Psi$, $\delta\Delta$) show measurement uncertainties in (Ψ, Δ) [1] and the subscripts *exp* and *mod* denote experimental and modeled, respectively, parameters at *j*th spectral point (*j*th photon energy E_j), and n is the number of spectral points.

As optimization tool, our HGGA algorithm is applied. The details about this algorithm may be found elsewhere [2]. Its main feature is the combination of the concept of genetic algorithm (GA) [9, 10] with a gradient-based search method to ensure a compromise between exploration of the whole search space and exploitation of the promising regions. The benefit is, in contrary to standard gradient-based methods like the ellipsometry traditional Levenberg-Marquardt, that the HGGA does not need any starting values for the model parameters to initialize the search process. More strictly speaking, the initial values of the model parameters are randomly generated by the algorithm, and are not delivered by the algorithm user. Consequently, the user avoids tedious search for the "good" starting parameters, what is a key factor in the success of the ellipsometry data evaluation process when using gradient methods. At the same time, for the global-search HGGA method, the initial values of the search parameters have minor impact on the obtained results.

Two most common criteria, namely, *AIC* [11], and *BIC* [12], are used to evaluate the selected optical dispersion model. These are defined as:

$$AIC = n\ln\left(\frac{RMSE}{n}\right)+2m \quad (2)$$

$$BIC = n\ln\left(\frac{RMSE}{n}\right)+m\ln(n) \quad (3)$$

where m denotes a number of model parameters. The "best" evaluated dispersion model is the one with the lowest IC value [5]. Thus, the aim of the analysis of experimental ellipsometry data implemented here is not only to minimize the *RMSE*, but also to find the model with minimal IC value. Consequently, the IC parameter can be assigned as a second criterion for the optimization process.

2.1 Evaluation of the method

For the verification of the proposed method, we used the ellipsometric spectra for diamond-like carbon (DLC) coating deposited on SW7M stainless steel, measured with the use of a Woollam M2000 ellipsometer in the photon energy range 1.2–5 eV, and for 55°, 65°, and 75° angles of incidence. Details about the sample preparation can be found in ref. [13]. It is known that the optical parameters of DLC strongly depended on the deposition method and process conditions [14–16]. Therefore, the reported reference data are varying from one sample to another. As a result, there is no good reference data for DLC available. The experimental spectra are shown as symbols in Figure 1. Interference fringes in the spectral range about 1.2–2 eV

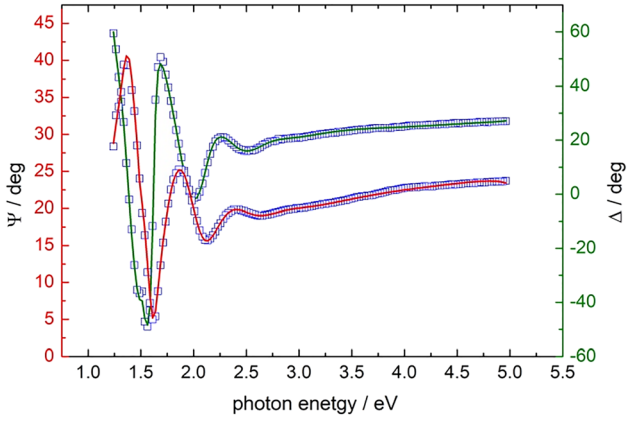

Figure 1: Experimental Ψ(E), Δ(*E*) spectra for DLC coating (only angle of incidence 75° is plotted for better clarity) – symbols, and modeled by the "best" dielectric function model (3 Gaussian oscillators) – solids.

clearly indicate the transparency regime of the measured DLC layer, while the absorption at higher energies needs to be taken into account when searching for the dielectric function model.

For the ellipsometry data analyzes, we assumed the so-called 4-phase optical model which consist of a semi-infinite substrate with known optical constants and a subsequent layer, whose optical properties have to be determined, followed by the surface roughness layer described by Bruggemann effective medium approximation (BEMA), assuming 50% of void [17]. For the description of the DLC layer optical constants, classical Lorentz and Gaussian oscillator functions were chosen [18]. Additionally, the Tauc-Lorentz (T-L) and the Cody-Lorentz (C-L) dispersion functions are evaluated [18]. The search parameters are the thicknesses of the DLC d_{DLC} and the surface roughness d_{r}, and the parameters of individual oscillators. Thus, the number of search parameters is varying in accordance to the complexity (number of oscillators) of the dispersion model used to describe the DLC optical constants.

First, we have analyzed the scenario where the complexity of the dispersion model is optimized in parallel with the exploration of the search space of model parameters. In this situation, the results of fitting were evaluated using three objective functions, namely the dependence of *RMS*, *AIC*, and *BIC* on the model parameters values and its complexity. The goal of such a multi-objective optimization was to find minimum values of all three criteria. This approach, however, leads to several major implementation problems which are connected with the varying number of search parameters during the optimization process. The search algorithm would need very large computation power. Convergence of the optimization process is also problematic, in particular for more complex models. Therefore, we decided to follow a more "standard" path, where at first step the search algorithm is started for each individual dispersion model. Then, the *AIC* and *BIC* are calculated for each solution and finally the ranking of the models is created (see Figure 2).

In Table 1, the obtained solutions are shown. We have analyzed 15 dispersion models in total: T-L, and C-L models as well as Lorentz (#L), and Gaussian (#G) oscillator models with different oscillator number, as denoted by the #. It should be emphasized that here the philosophy is to obtain "good" model match – avoiding under- or overfitting, rather than the physical meaning of the model itself. More strictly speaking, relying only on the statistical criteria at this stage, intentionally omitting subjective in nature assessment by a human expert. Nevertheless, certain boundaries for the variation space of the model parameters are necessary. First, the oscillator strength and width parameters values need to be positive. Next, the oscillator shall be placed in the analyzed spectral range or at least in reasonable neighborhood of it. Here we assumed that the oscillator central energy parameter value cannot exceed the photon energy range 0–20 eV.

After all these assumptions, the HGGA search algorithm converge to the minimum value of *RMSE* even for the most complex dispersion model – here 7 Lorentz oscillators, usually after about 30 generations. The number of candidate solution at each iteration (population size) of the HGGA and the crossover fraction parameter of the algorithm (see Ref. [2] for details) was set as 2000 and 0.5, respectively, for each algorithm run. An example convergence curve is shown in Figure 3. As the algorithm is based partly on the stochastic process [2], rerun of the optimization process is sometimes needed before reaching the global minimum in the search space. To compare the evaluated models more easily, IC differences *dAIC* and *dBIC* are also calculated together with raw *AIC* and *BIC*:

$$dIC_k = IC_k - IC_{\min}, \qquad k = 1, \ldots, q, \qquad [4]$$

where k is the number of candidate models under test, $IC_{\min}$ is the score of the "best" candidate model [5]. This

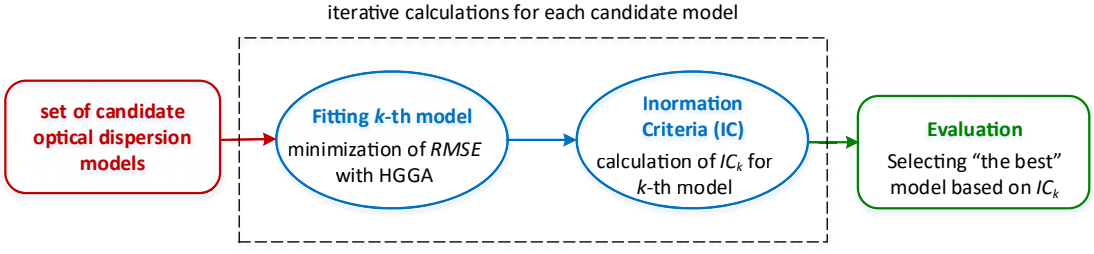

Figure 2: General procedure flowchart illustrating the major steps of the selecting "the best" model over the candidates with the method proposed here.

Table 1: Ranking of the evaluated dielectric function models. Supported models are denoted by bold.

	C-L	T-L	1L	2L	3L	4L	5L	6L	7L	1G	2G	3G	4G	5G	6G
AIC	480.40	465.11	527.36	440.39	420.00	423.74	430.17	454.76	423.78	477.20	426.10	418.07	423.42	418.84	428.41
BIC	501.52	477.18	539.42	461.51	450.17	462.97	478.45	512.09	484.13	489.27	447.22	448.24	462.64	467.12	485.74
dAIC	62.34	47.05	109.29	22.32	1.93	5.68	12.11	36.70	5.72	59.13	8.04	0.00	5.35	0.78	10.35
dBIC	54.30	29.96	92.90	14.28	2.95	15.74	31.23	64.87	36.90	42.04	0.00	1.01	15.42	19.89	38.52
RMSE	21.95	20.64	31.17	16.84	14.14	13.93	13.97	12.78	12.7	22.36	14.22	14.13	13.11	13.36	13.27

parameter is showing "how far" is the kth model from the "best" one. Following [5], the models with $dIC_k < 2$ are considered almost as good as "best" model, while those with $dIC_k > 10$ are very unlikely.

To visualize the obtained results, the dependency of the dIC_k with the complexity of the model is plotted in Figure 4. Gray rectangle denotes the IC_k values below 2. According to *dAIC* criterion, the "best" model is that with 3 Gaussian oscillators (3G). Other models supported by *dAIC* are 3L and 5G models with *dAIC* values 1.93 and 0.78, respectively. The 3G model is also supported by *dBIC* criterion (*dBIC* = 1.01), but slightly better *dBIC* value is obtained for 2G model. However, taking into account that for 2G model *dAIC* criterion value exceeds 2, and for 3G model both criteria are in the "supporting region", the model with 3 Gaussian oscillators can be treated as the "best" among the evaluated models. On the other hand, it is clearly seen that C-L and T-L models, which are usually used to describe the dielectric function of DLC, are much less advisable for this particular case. The dielectric function obtained by the "winning" model is shown in Figure 5. The determined thicknesses of DLC and roughness layers are 588 and 8.8 nm, respectively.

It can be also noticed in Table 1 that models with 3 and more Lorentz oscillators and likewise two and more Gaussian oscillators possess similar *RMSE* values – for more complex models *RMSE* is even slightly lower. Hence, the dielectric functions derived with these models are basically the same when judging "by the look". Nevertheless, the IC penalize more complex models as the increasing of the number of model parameters do not have serious impact on the fitting result in this case. Consequently, the applying of models with larger number of oscillators leads to the overestimation. This clearly shows that searching only for "as

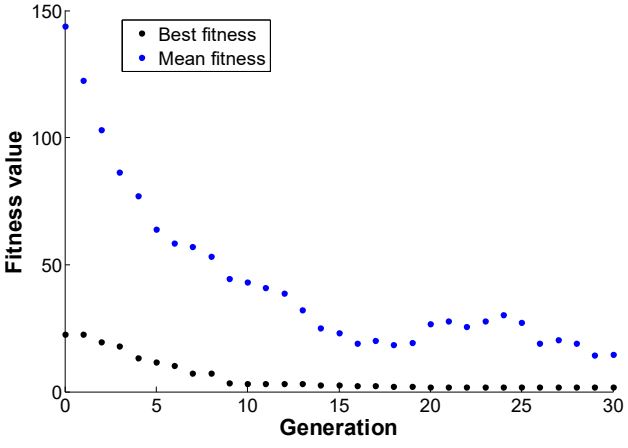

Figure 3: An example HGGA convergence curve for the DLC coating on SW7M.

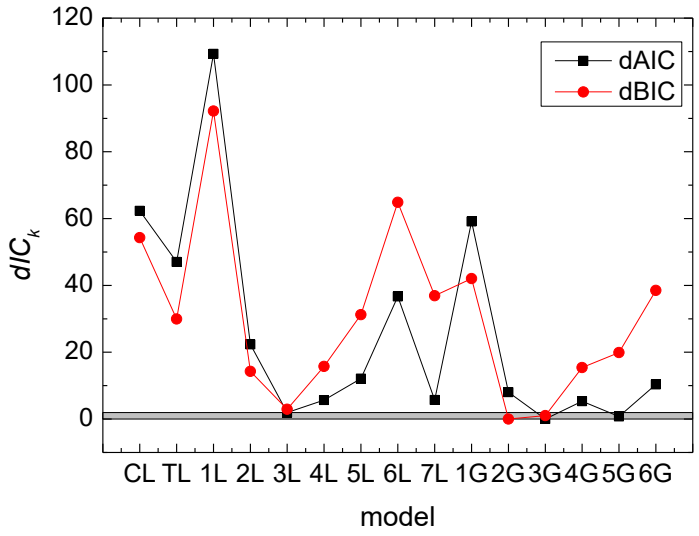

Figure 4: The variation of the information criteria with the complexity of the dielectric function model. Gray area indicates the supported models according to their IC value.

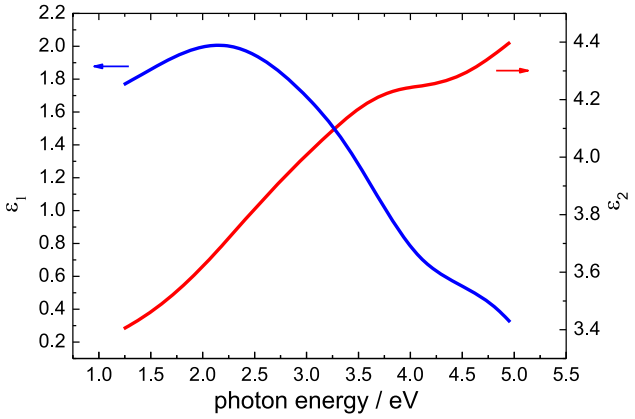

Figure 5: Dielectric function for the DLC coating (thickness 588 nm) deposited on SW7M stainless steel derived from the "best" model (3 Gaussian oscillators).

low as possible" value of *RMSE* should not be the only aim of ellipsometry data evaluation.

3 Conclusions

In this work, we demonstrated the capabilities of a search algorithm which applies the concept of genetic algorithm to find the optical model parameters in the evaluation of experimental ellipsometric data. Compared to the standard for ellipsometry gradient-based algorithms, the method shown here does not need any starting values delivered by the user to initialize the search process. Additionally, as a global-search method, it allows to explore a large area of the search space. Therefore, the proposed algorithm is particularly attractive for the ellipsometric data evaluation in the case of limited *a-priori* knowledge about the sample under studies. Due to their local search approach, typical gradient optimization methods require tedious search for good starting points which are close enough to the actual solution.

Employing another optimization criteria, namely the statistical information criteria, as a next to the standard root means square error optimization criterion, allows to explore not only the model search parameter values space but also the space of different dispersion models used to describe the unknown optical parameters of the model. As a result, our combination of the global-search algorithm with the statistical information criteria seems a powerful method for unambiguous selection of the most appropriate dielectric function model (within a given set of models) for the description of the optical constants of the studied material. Finally, as the method allows for the complete formalization and automatization of the ellipsometry data analysis process, it can be attractive for the use by either non-experienced or commercial users.

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Conflict of interest statement: The authors declare no conflicts of interest regarding this article.

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