

# THE LIMITATIONS OF SOLUTION ACCURACY FOR ELLIPSOMETRIC PROBLEMS

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UDC 535.51;39

Contemporary nanotechnologies often require calculating of three parameters of metal films simultaneously using ellipsometric measurements at single wavelength. As a rule these measurements are needed for routine control of deposition conditions. The knowledge of exact values of film thickness and complex refractive index is highly important because they define reliability of device operation. In this paper a new algorithm of inverse ellipsometric problem solution using Mathcad platform is proposed and difficulties in obtaining accurate results are considered. The solution accuracy limiting factors are the following: measurement accuracy, quality of ellipsometer alignment and homogeneity or uniformity of object of study.

**Keywords:** ellipsometry, optical properties, inverse ellipsometric problem, transition metal, monitoring process.

## Introduction

Ellipsometry is a nondestructive and no-disturbing method of measuring of properties of both dielectric and metallic films. Advantage of Ellipsometry method among different optical techniques consists in that both amplitude and phase of electromagnetic wave which are reflected from surface of films are extracted simultaneously when every measurement. Specific difficulties are appeared in case of strongly absorbing films, for examples, metals and some semiconductors. Aluminum films, films of most transition metals (such as Titanium, Vanadium, Magnesium, Cobalt, Nickel, and Palladium), noble metal films (such as gold and silver films) and copper films and their oxides are widely used in different micro- and nano-devices. It is well known that film morphology and film composition depend on preparation technology. Unfortunately, nomograms method of definition of the three parameters of absorbing films is inapplicable in this case. Author repeatedly focused their efforts on development of new approaches to this difficult problem of determination of properties of different materials [1, 2, 3, 4].

## 1 Principal questions

### 1.1 Survey of Methods of Film Parameters Reconstruction Based on Ellipsometric Measurements

#### 1.1.1 Nomograms Method

Nomograms method was proposed by Archer [5] in 1962 and improved by Saxena [6] three years later, and for many years it was the only method for an interpretation ellipsometric measurements. This method is used for determination of the properties of transparent films (silicon dioxide or silicon nitride, as a rule) on silicon substrate; lines of equal film thicknesses together with lines of equal refractive indices are displayed on plane of ellipsometric angles  $\Psi$  and  $\Delta$  for single wavelength and single angle of incidence of light. At first, nomograms were drawn on paper sheets stitched into an album, later personal computers allowed us to display film parameters quickly using monitor screen.

#### 1.1.2 Computer Methods of solution of inverse ellipsometric problem (IEP)

First of all it is very useful to read paragraph 4.5 titled "Numerical inversion of the exact equation of reflection ellipsometry" in the book of R.M.A. Azzam and N.M. Bashara [7] where some aspects of IEP solutions such as:

cases of analytical solution, choice of error function ( $E_r$ ), rate of convergence for  $E_r$  minimization, and hence, choice of angles of incidence, and in addition affect of experimental errors were considered. It is absolutely clear that there is no standard solution, and every task demands particular approaches.

### 1.1.3 Transparent Films

The solution for transparent film is the easiest case. But keep in your mind that the possibility of finding false solution still exists if the structure being analyzed consists of double dielectric layers, for instance, as in ONO (oxide-nitride-oxide)-Structure [4, 8]. The accuracy could be increased by some times when statistical processing of found solutions of inverse ellipsometric problem was done, so that to avoid finding local minimums as in [2]. Furthermore, purely mathematical specifics of determining of accurate solution were analyzed: conditionality, uniqueness, stability, and mutual dependence of solution IEP [2, 8].

### 1.1.4 Absorbing Films

Earlier in [1], technique of finding a great number of exact solutions using one pair of ellipsometric angles was developed, but additional measurement should be taken for removal of ambiguity. The solution could be obtained in form of space (three-dimensional: n-k-d) spiral, every point of which corresponds to exact solution of ellipsometric equation. The point of intersection of two three-dimensional spirals gives us the only solution. By such way, map of film parameters (thickness, refractive index, and absorption coefficient) was created for film of mercury telluride HgTe [4]. Besides, the analysis of general ambiguity of IEP was carried out and existence of several families of IEP solutions was revealed [3] as shown on different experimental examples. All above mentioned reasoning relate to measurements at single wavelength.

### 1.1.5 Spectroscopic ellipsometric measurements

Spectroscopic ellipsometry requires both particular modeling and dividing of task into two or more sub-tasks so that two parts of specter — transparency region and region of strong absorption — could be used for successful IEP solution, for instance as in [8, 9, 10]. Building of Adequate Model of film structures provides reliability of result of EIP-solution. Model has to take into account that film properties differ from its bulk properties. It needs to use Lorentz's oscillator's model for description of permittivity of each film when fitting procedure is applied, i.e. getting one-to-one correspondence of experimental data and calculations for mathematic model, using method of least-squares (such as Levenberg–Marquardt algorithm).

## 2 Experiment

### 2.1 Equipment

Measurements of optical constants and thicknesses of metallic films are important now and they will be vital question along with other questions of technology and both mathematic description and modeling. An accurate knowledge of all properties of films is needed for desired functionality of device. In our experiments, film depositions were carried out in high vacuum unit with (ALD) technique of additional layer deposition. LEP-3M (laser ellipsometer photometric) was used for ellipsometric measurements at wavelength of 633 nm.

### 2.2 Programming

Calculations of parameters of metallic films were carried out using the Mathcad 15 platform keeping in mind all its advantages and demerits which are listed below.

The benefits of programming using Mathcad are the following:

1. Fast and easy to write a program (you do not need knowledge of algorithmic languages).
2. Fast debugging because you can display anywhere in a program the results of intermediate calculations.
3. Quick and convenient reading data from a file and record the calculated data in the file.

Disadvantages of programming using Mathcad are the following:

1. Slow (by 1-2 orders of computation speed magnitude inferior to similar programs written using high-level languages like FORTRAN and PASCAL).
2. Lack of flexibility of the programming language.
3. No possibility to compile and link programs and, hence, run it separately from the main program block.

### 2.3 Calculation and Algorithm of Finding Solutions

All calculations are based on Equations which earlier had been gotten in [1]: according to [8], ellipsometric Equation for reflection coefficient of polarized light can be written by Eq. (1), where the only  $X$  is unknown variable. Multipliers of square equations  $A$ ,  $B$ ,  $C$ ,  $D$ ,  $E$ , and  $F$  are cumbersome combinations of Fresnel reflection coefficients.

$$\rho \equiv \tan \Psi e^{i\Delta} = \frac{A + BX + CX^2}{D + EX + FX^2}, \quad \text{where } X = \exp \left[ -\frac{4\pi d}{\lambda} (\gamma + i\beta) \right], \quad (1)$$

The roots of Equation (1) are two complex values  $X_1$  and  $X_2$ :

$$X_{1,2} = \frac{-(B - \rho E) \pm \sqrt{(B - \rho E)^2 - 4(C - \rho F)(A - \rho D)}}{2(C - \rho F)} = |X_{1,2}| e^{i\theta_{1,2}}. \quad (2)$$

It is easy to show that the real and imaginary parts of  $X_1$  and  $X_2$  satisfy the equation of the logarithmic spiral:

$$-\frac{1}{\gamma} |X_{1,2}| = \frac{1}{\beta} \theta_{1,2}. \quad (3)$$

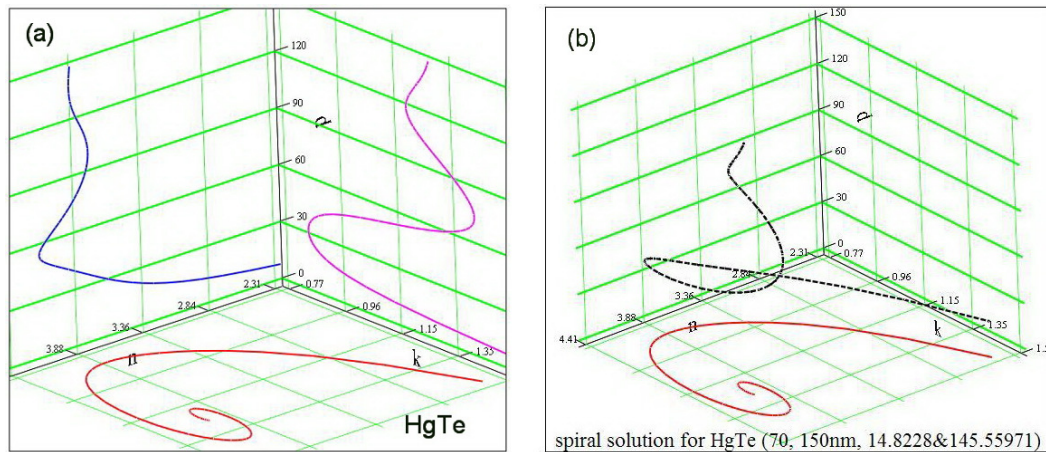


Figure 1: A full set of exact solutions in 3d-space  $n$ - $k$ - $d$  for single pair of ellipsometric angles  $\Psi = 14.822$  and  $\Delta = 145.55971$  measured at one incidence angle of light beam  $\varphi_0 = 70^\circ$  for mercury telluride HgTe film with thickness of 150 nm,  $\lambda = 632.8$  nm. (a)-Three projections ( $n$ - $k$ ,  $n$ - $d$ ,  $k$ - $d$  and (b)-3D spiral.

You can see it for mercury telluride in Figure 1. Analogical three projections and general solution of spiral-curve for transition metal such as palladium are demonstrated in Figure 2, unfortunately, they are less expressive than former 3-D graphic.

Values included in equation 3 do not depend on thickness  $d$ , so, this projection can be built on the plane  $n$ - $k$ . Two another projections of the general solution  $n$ - $d$ , and  $k$ - $d$  are also shown in Figure 1; the first projection has the form of distorted (deformed) logarithmic spiral due to non-linearity transformation  $X \rightarrow N = n - ik$ . The three-dimensional spirals are shown in the right part of Figure 1 and of Figure 2 together with the  $n - k$  projection for two materials: mercury telluride -HgTe and Palladium, respectively.

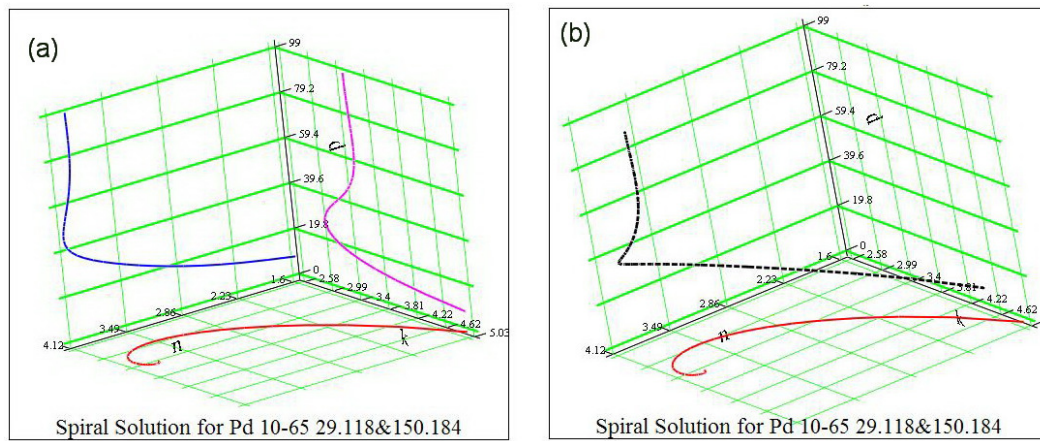


Figure 2: A full set of exact solutions in 3d-space  $n$ - $k$ - $d$  for single pair of ellipsometric angles  $\Psi = 29.118$  and  $\Delta = 150.18398$  measured at one incidence angle of light beam  $\varphi_0 = 65^\circ$  for palladium Pd film with thickness of 10 nm.  $\lambda = 616.9$  nm,  $N_{Si} = 3.89903 - 0.00753i$ ,  $N_{Pd} = 1.75 - 4.21i$ .

### 2.3.1 Elimination of an ambiguity to get exact Equations solution

The exact solution of the main ellipsometric equation for finding optical constants and thickness of thin ( $\sim 10$  nm) metal films is an essential but difficult task. Multiple-angle-of-incidence (MAI)-ellipsometry could be also successful in case of absorbing films, if algorithm of calculations will be improved. Application of equations (1-3) led us to a method which could be named as analytical method of spatial-spiral or of three-dimensional spiral.

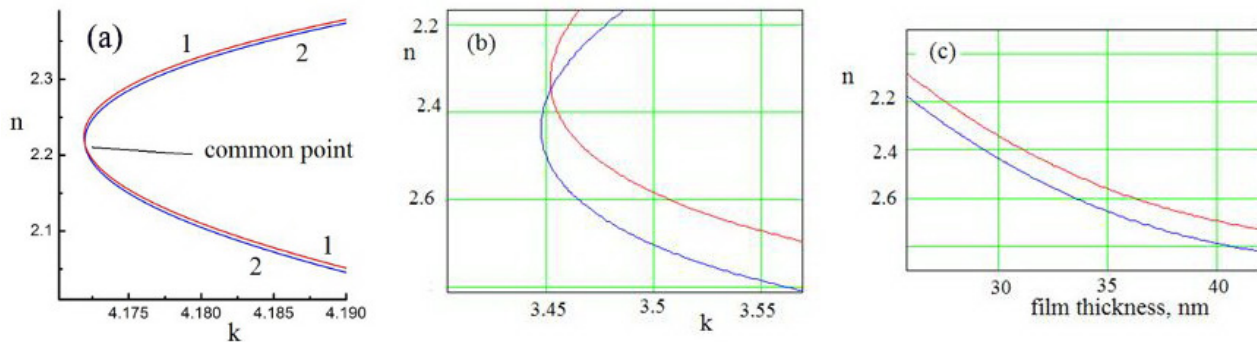


Figure 3: Common point from TOTAL SET of EXACT SOLUTIONS in 2D-spaces for two pairs of ellipsometric angles  $\Psi$  and  $\Delta$  measured at TWO ANGLES of incidence of light beam  $\varphi_{01}$  and  $\varphi_{02}$  for TITANIUM films: (a)-for simulated data, (b) and (c)-for experimental data.

The main idea of the method is the following. Measurements were carried out at a number of different angles of incidence of light beam ( $\varphi_{01}, \dots, \varphi_{0m}$ ) and  $m$ -pairs of ellipsometric angles were obtained ( $\Psi_1, \Delta_1, \dots, \Psi_m, \Delta_m$ ). All possible exact solutions were found using equations (1-3) for every  $\varphi_{0i}$  in form of spatial-spiral. Since, all spatial-spirals should have only one common point of intersection which corresponds to exact solution for metallic film, as shown in Figure 3. **The definition of this intersection point is the aim of proposed algorithm.** The IEP should be solved step-by-step for every value of film thickness from range which contains the desired thickness; starting from maximal thickness in this range. Complex refractive index of this thick film will be close to the same index as in case of semi-infinite medium. And besides, the IEP should be independently solved for two different angles of incidence of light beam ( $\varphi_{01}, \varphi_{02}$ ) to get the common intersection point of two spirals. So, curves of solutions do not contain breaks or discontinuities, therefore, every next point could be defined using value of previous point. In practice, intersection point in three-dimensional space ( $n_f - k_f - d_f$ ) of film parameters can be lacking, due to inadequacy of model of description of structure or measurement's errors. In this case, it is possible to define intersection point in two-dimensional space, i.e. at plane ( $n_f - k_f$ ) for two values of angles of incidence of light beam. Notice that the values of film thickness in this point will be different  $d_f(\varphi_{01}) \neq d_f(\varphi_{02})$ ,

and the smaller their discrepancy than more accurate solution. Hence, some *approximate* solution could be defined if desired values belong to permissible intervals.

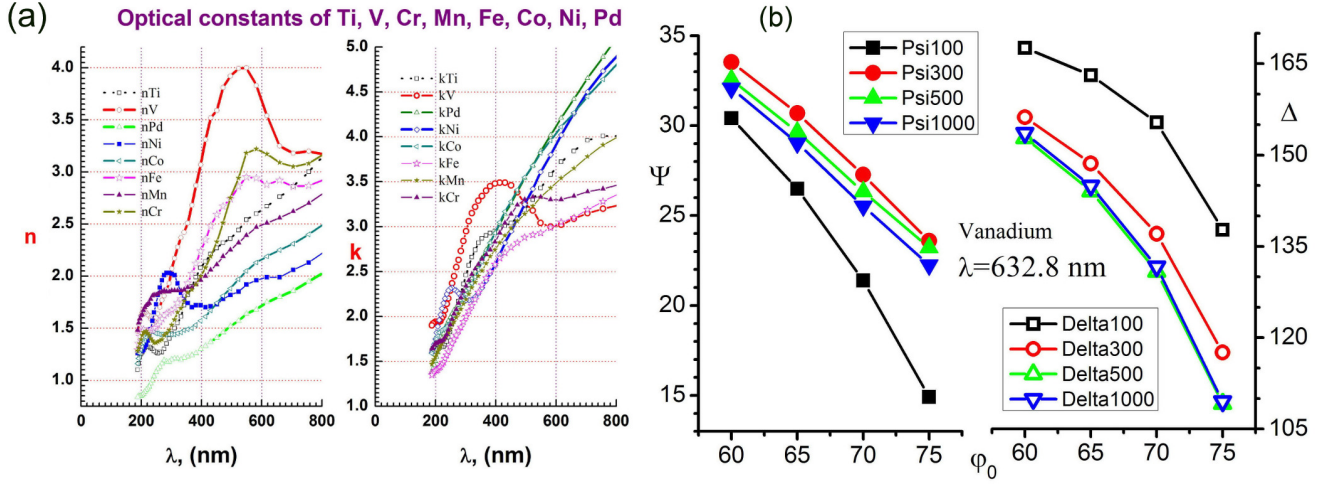


Figure 4: The optical properties of transition metals as in paper [11]; and Ellipsometric angles  $\Psi$  and  $\Delta$  via angle incidence of light beam  $\phi_0$  and film thickness (in nanometers) for Vanadium film with various thicknesses.

### 3 Ellipsometric Angles as functions of Parameters of Metallic Films

In this section reasons of appearance of measurement errors will be investigated. Let us suggest that erroneous measurements are only introduced by object, i.e. by metallic film; and analysis of the nature of such errors will be represented. The main question: is it possible to avoid experimental errors which do not allow getting exact solution? To answer that question, a number of metal and of their optical properties had been considered. We paid attention two groups of metals: transition metals and noble metals. All metals included in these groups exhibit strong absorption of light at wavelength 633 nm. Therefore, for the following steps of reconstruction of film parameters, real experiment's measurements were substituted on simulation Data obtained using evaluation of ellipsometric angles  $\Psi$  and  $\Delta$  for all materials for set of  $\phi_0 \in 60^\circ, 65^\circ, 70^\circ, 75^\circ$  and for a set of film thicknesses (10, 30, 50, 100 nm). Simulation was applied to avoid any errors. Figure 4 demonstrates optical properties of transition metal, as for instance; and variations of polarization angles of vanadium. The changes of ellipsometric angles  $\Psi$  and  $\Delta$  become smaller with increasing film thickness, as shown in Figure 4. Hence, quality of IEP estimation only depends upon both algorithm of program and optical properties of metal. Above described algorithm of program was used in evaluation for the whole set of simulated ellipsometric angles. Besides, noble metals (Au, Ag, and Cu) were used for testing of proposed algorithm. The analysis revealed that estimations of the desired film parameters ( $n$ ,  $k$ ,  $d$ ) were received with high accuracy in the range of conditions of simulation data. However, successful IEP solution requires high precision of input Data, so ellipsometric angles should be known with accuracy up to four decimal places (i.e.  $0.0001^\circ$ ), as for Ti film, for instance. It is well known that technological conditions of film deposition could hardly provide this accuracy and, mainly, due to the inhomogeneity of the composition and structure of thin films. It is super difficult task. Precision of goniometer devices in ellipsometer could be increased but the effect of this increase would be extinguished due to variability of film properties under deposition. There are other serious obstacles related to accuracy of definition of thin film thickness in particular. Details of impact of absorption on polarization of reflected light are displayed in Figure 4, for example, for Vanadium. Vanadium film was deposited on Silicon substrate and measurement were carried out at wavelength  $\lambda = 633$  nm. Notice that Vanadium film thickness of 100 nm exceeded thickness period, i.e. film phase thickness is equal or more than  $2\pi$  therefore its curve (100 nm) is located below previous curves. Obviously, exact solutions could be found for film thickness which is equal to 10 nm or smaller.

## Summary

Parameters of thin (nano-scale) metal films could be defined by ellipsometric method using proposed program algorithm of construction of three-dimensional exact solution. Application of Mathcad platform for implementation of this algorithm gives us possibility to study nuances of calculation procedure and to reveal restrictions preventing to get exact solution.

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Received — May 24, 2017*