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NUMERICAL ANALYSIS OF ZNO THIN LAYERS HAVING ROUGH SURFACE

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Abstract. *In this paper an automated procedure for the analysis of Transparent Conductive Oxides (TCO) layers exhibiting rough surfaces is proposed. The method is based on the interaction between MATLAB and the Sentaurus TCAD and is aimed to the reduction of computational efforts needed for full three dimensional analyses. Experiments performed on CVD deposited ZnO layer, showing the reliability of the method for describing their optical properties, are reported. A semi-empirical technique for the extraction of the TCO refractive index is shown as well.*

Key words: *TCO, AFM, ZnO, Refractive index*

1. INTRODUCTION

The characterization of Transparent and Conductive Oxides (TCO) is an open task for the scientific community. This is mainly due to the fact that in many applications, as an example when they are exploited as anti reflective coatings in optoelectronic devices, they exhibit very rough surfaces and conventional one dimensional models are unreliable to effectively describe their behavior. Many advanced software packages allow full three dimensional capabilities for accurate numerical simulations of arbitrarily shaped surfaces but, as they operate on a discretized mesh which reproduces the real surface, the number of required grid points is often too large, leading to unpractical computational time. A second issue depends on the unreliability of geometrical models for modeling light propagation when roughness induces diffraction effects.

In this paper we propose an automated procedure, based on the interaction between MATLAB and Sentaurus TCAD [1,2], which finds, in a generic surface, the smallest area characterized by the same statistical features (average roughness, standard deviation and so on) of the whole surface; thus, a reduced device, with same optical properties of the

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real one, can be defined and analyzed. The same procedure looks for the existence of a two dimensional section where statistical parameters are conserved as well, so that, in some cases, the analysis can be reduced to a two dimensional one. The availability of this procedure allowed us to check the limits of light propagation models for a given structure, and lead us to define whether exact solution of Maxwell equations are needed to take into account diffraction phenomena [3,4].

Moreover, the availability of a 3D model allowed us to set a semi empirical procedure to achieve the wavelength dependent refractive index of thin ZnO layers. It should be underlined that our aim was to look for a global index able to describe light intensity transmitted beyond a given TCO film which, independently of its actual physical meaning, allows reliable numerical simulations of optoelectronic devices.

Many samples were specifically fabricated by varying deposition parameters to achieve different surface roughness (from smooth to very rough). Comparison between experiments and simulations, the latter performed by importing real device geometries, proved the reliability of our approach.

The paper is organized as follows. In Section II the MATLAB code which evaluates statistical parameters and looks for the minimal surface is described. In Section III the procedure is applied to experimental samples made by CVD deposited ZnO thin film [5,6]. Section IV describes the procedure for evaluating the complex refraction index of deposited layers. Conclusions are drawn in Section V.

2. Processing of AFM Images

As previously mentioned all layers analyzed in this paper were CVD deposited ZnO thin films. Fig.1 shows an example chosen among the Atomic Force Microscopy (AFM) images gained for our samples.

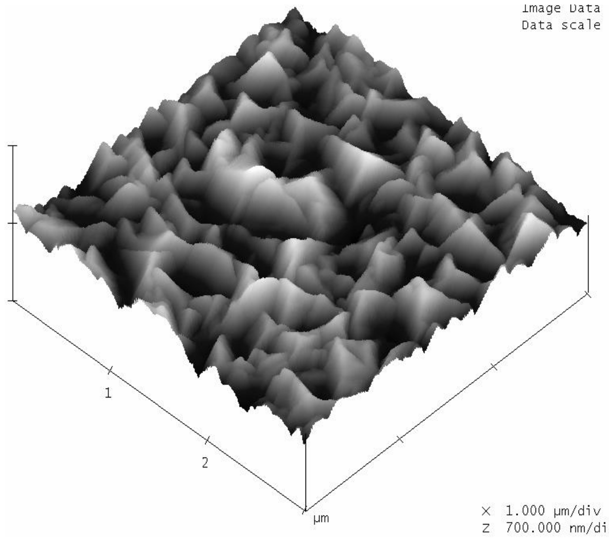

Fig. 1 AFM image of a CVD deposited ZnO thin film

As can be seen pyramidal shapes are randomatically distributed over the surface, thus assuring an antireflective behavior to the film.

As first step of the processing the AFM images were loaded in the MATLAB environment. A Graphical User Interface (GUI), shown in Fig. 2, was built for a user friendly managing of the files. The GUI allows commands input and results visualization; as an example, in the upper right corner of Fig.2 we can see the image of Fig.1 reproduced as MATLAB plot

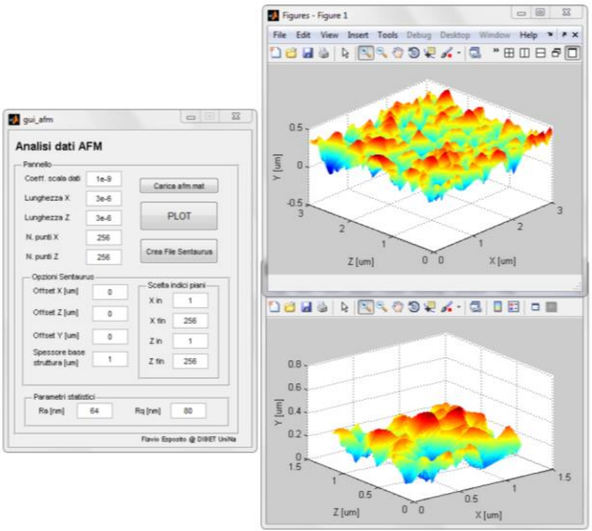

Fig. 2 MATLAB GUI interface

The MATLAB code underlying the GUI has three main features, it evaluates statistical roughness parameter of the whole image; looks for the smallest surface with same parameters; automatically generates the Sentaurus input file for numerical analysis.

Statistical parameters are evaluated according to the definition of the effective roughness height given in [7]

$$\sigma_{rms} = \sqrt{\frac{1}{S}\int_S r^2(x,y)dxdy} \tag{1}$$

where *r* is the difference between the profile eight and its mean value and S is the area of the sample. The parameter given in (1) is iteratively evaluated by considering decreasing portions of the total area by means of a successive halving criterion. The algorithm stops when the smallest area still holding the starting value of the effective roughness is found. Then, the algorithm verifies that different portions of the total surface having the same area just defined exhibit same parameters, thus assuring the statistical robustness of the procedure. Once a reduced surface is chosen (an example is shown in the low right corner of Fig.2) the three dimensional analysis of the properties of the thin layer, with respect to light propagation, can be effectively performed by exploiting a corresponding reduced set of grid points. For the example shown in Fig.2 we achieved a reduction of grid points of about 75% and a reduction of the computational time which was greater than 90%. With the aim of further reducing computational efforts the MATLAB code gives the chance to verify if the analysis of the surface properties can be reduced to a two dimensional problem. In other words, the effective roughness defined by (1) is evaluated along a finite set of transversal cross sections (the step of the scan is adjustable from the GUI). If the effective roughness for the cross section is the same already valuated for the reduced surface the subsequent numerical analysis is performed in a two-dimensional reference system, otherwise, full three-dimensional analysis is performed. The procedure ends after automatically generating the input code for the Sentaurus environment.

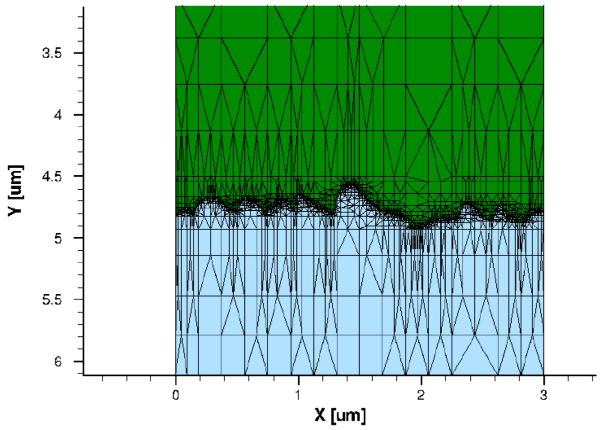

Fig. 3 Sentaurus 2D mesh for the cross section of an AFM image

As an example Fig.3 shows an image of the 2D grid generated by Sentaurus after receiving the input from MATLAB, while Fig.4 shows the analogous for a 3D case.

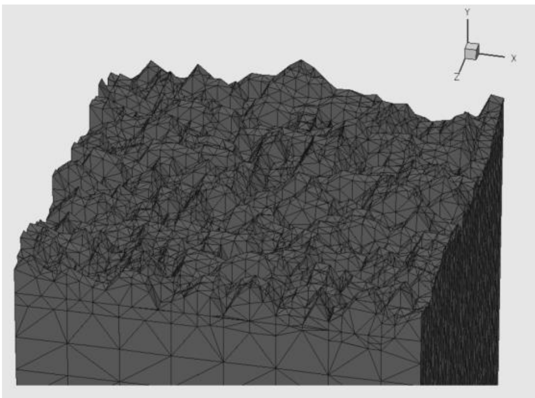

Fig. 4 Three dimensional Sentaurus mesh for an AFM image

Once statistical parameters have been determined a further simplification, which can eventually be adopted, consists in the substitution of the real surface with an equivalent surface only formed by regular pyramids. Geometries of the pyramids should be chosen so as to have same statistical parameters of the real surface. The reliability of this simplification is evidenced in Fig.5 and Fig.6.

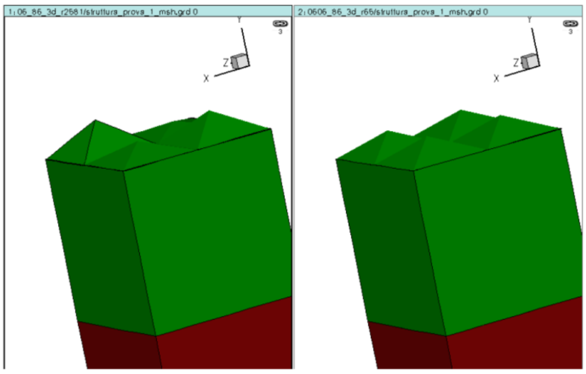

Fig. 5 Sentaurus structure with non uniform (left) and uniform (right) pyramids

The structure in the left side of Fig.5 has pyramids with different heights while the structure in the right side has uniform pyramids but their heights are chosen to have same statistical parameters of the non uniform structure.

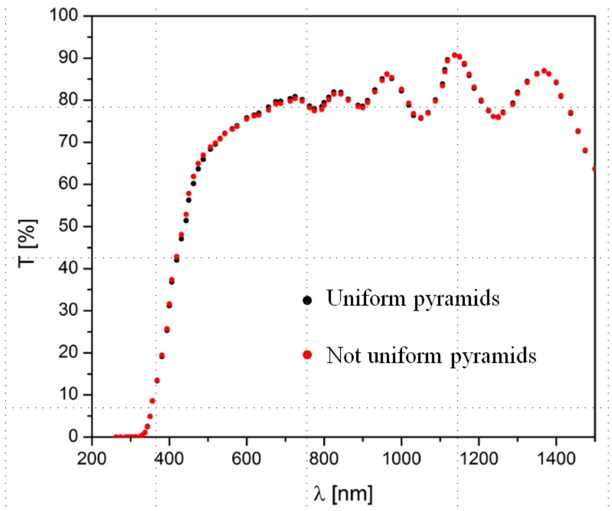

Fig. 6 Transmittances profiles evaluated by Sentaurus for structures with different surface shape but same effective roughness.

As can be seen in Fig. 6 the transmittances evaluated for the two cases are perfectly coincident. This fact means that, in principle, optical properties of a real surface, like that of Fig.1, can be reliably analyzed by considering a corresponding simplified surface, once statistical parameters have been evaluated.

3. Experiments

All deposited ZnO films were both optically and electrically characterized, in particular optical properties were defined in terms of transmittance profiles. Experimental transmittances were then compared with those numerically evaluated by Sentaurus on the previously defined reduced surfaces. Sentaurus makes available three models for describing both light propagation through materials of given optical properties and light interaction with interfaces between materials with different optical properties. The simplest one is the TMM (Transfer Matrix Method) model [8] which only applies to flat surfaces so that it is not suitable for our cases. The second model considers light as formed by discrete "rays" and applies geometric optics laws for reflection and transmission. Usually, this method is preferred over others because of the reduced computational efforts. However it should be considered that diffraction effects may not be negligible because the width of pyramids like those shown in Fig.1 can be comparable with the wavelengths in the visible spectrum. In such cases the exact solution of Maxwell equations should be evaluated; the special model allowed for this application is termed EMW (Electro Magnetic Wave solver) in the Sentaurus environment. The reliability of both Ray-tracing and EMW models were checked with reference to samples either subjected or not subjected to diffraction effects. As an example Fig. 7 shows the comparison between the measured transmittance and the corresponding profile evaluated by means of the Ray-tracing method when no diffraction is present.

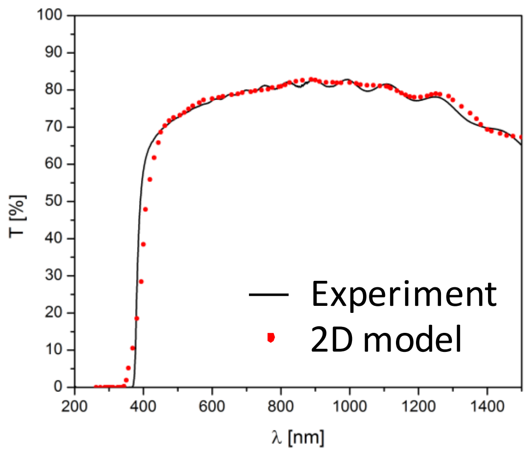

Fig. 7 Comparison between a measured transmittance and a numerical result achieved by means of the ray-tracing model.

Note that, in this case, according to the criteria defined in section II, a 2D analysis was performed. The agreement between the curves shown in Fig.7 evidences that the 2D analysis is enough accurate; moreover this result supports the reliability of the grid points reduction procedure described in the previous section.

An example of results gained by means of a 3D analysis is reported in Fig.8.

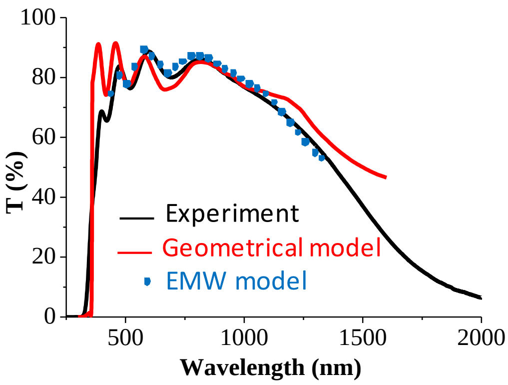

Fig. 8 Measured and numerical transmittance profiles for a surface encountering diffraction effects. The EMW model correctly reproduces the experimental curve.

The figure refers to a surface manifesting diffraction effects. As can be seen the transmittance profile achieved by means of the geometrical approach (ray-tracing) is a poor description of the measured transmittance. On the other hand, the transmittance profile gained by the EMW model correctly reproduces the experimental one. It should be emphasized again that the numerically evaluated transmittance profile was achieved on a reduced 3D surface automatically determined by the MATLAB code.

4. Refractive Index Measurements

The procedure which allows evaluating the transmittance profile once geometries and optical parameters of a given material are known can be conversely used for determining optical parameters of an unknown material once surface geometry and measured transmittance profiles are available.

The method described hereafter refers to the characterization of a ZnO layer deposited on a glass substrate (of assigned thickness and optical parameters) whose transmittance profile was previously measured for different angles of the incident light.

Only the refractive index of the ZnO TCO is assumed to be unknown.

The transmittance was first determined by Sentaurus for every possible value of both the real part *n* and the imaginary part *k* of the refractive index. Indeed, for a given structure, it is always possible to draw a surface like that shown in Fig.9, which, for an assigned wavelength, is the locus the transmittances compatible with all possible *n*,*k* couples.

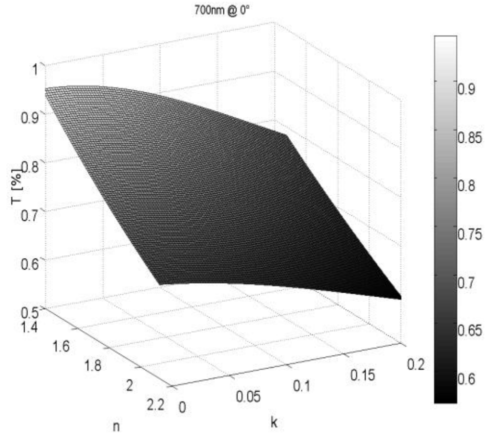

Fig. 9 Transmittance for a ZnO on glass structure as a function of the complex refractive index of the ZnO

The surface shown Fig.9 was achieved by assuming normal incidence (0°) of the light; it was compared with the transmittance actually measured in the same conditions which was 82%. The intercept of this value with the surface gives a curve in the plane *n*,*k*, as shown in Fig.10. All couples *n*,*k* belonging to the curve lead to the same value of the transmittance; from an applicative point of view this fact means that, in order to evaluate the light which is transmitted to an underlying device all that couples are equivalent and the actual refractive index is not needed. The above result only applies to normal incidence of the light. However, in real devices, the angle of incidence of the light is usually not known *a priori*, thus a refinement of the extraction is needed.

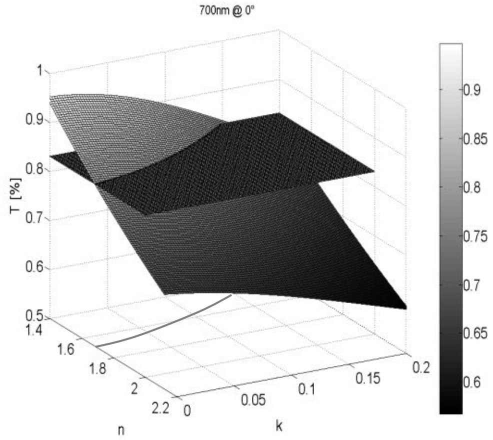

Fig. 10 Intercept between the transmittance surface and the measured transmittance. The line is the locus of all refractive indexes giving the same transmittance for the assigned ZnO on glass structure.

To this end we first considered a possible uncertainty affecting the measurement of the transmittance.

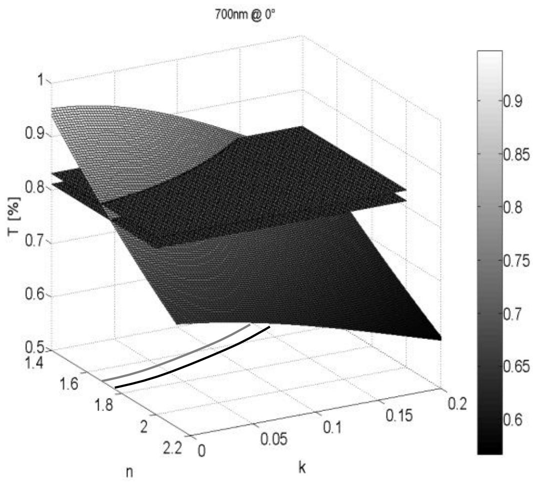

Fig. 11 Transmittance affected by a measurement error.

As an example in Fig.11 we considered that the measured transmittance was 82% +/- 1%. Therefore, a region of the *n*,*k* plane (instead of a curve), where refractive index compatible with the measured transmittance lie, was now identified. The portion of the n,k plane between the two lines is, indeed, the locus of refractive indexes giving the transmittances in the assigned range. Then, the transmittance surface was determined for a 15° incidence angle, as shown in Fig.12, and compared with the corresponding measured transmittance, (78%) +/- 1%, which is shown as well.

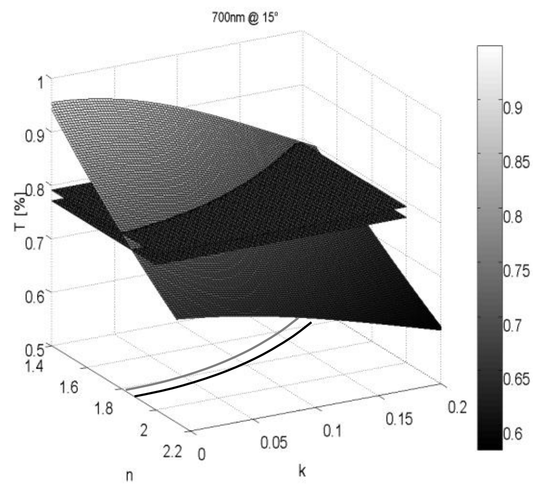

Fig. 12 Transmittance surface and measured transmittance for a 15° incidence angle.

A new region of possible n,k couples compatible with the measured transmittance was thus determined. As the TCO is always the same its actual refractive index, which is independent of the incidence angle, should give both measured transmittances. Hence the actual refractive index belongs to both regions identified in Fig.11 and Fig.12.

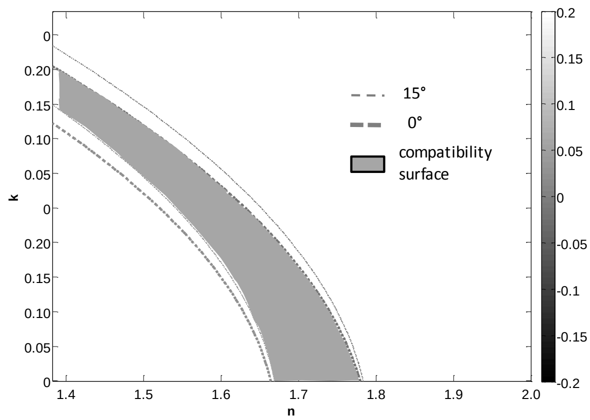

Fig. 13 Locus of refractive index giving the transmittance measured for both 0° and 15° incidence angle.

For the sake of clarity Fig.13 shows, in the plane n,k, the two regions identified in Fig.11 and Fig.12; points lying in the shaded region give both the transmittance measured at 0° and the transmittance measured at 15°.

The procedure can be iterated to further reduce the spread for the *n*,*k* values to be adopted for simulations. Fig.14 shows the result we gained by adding two further incidence angles, 45° and 60°.

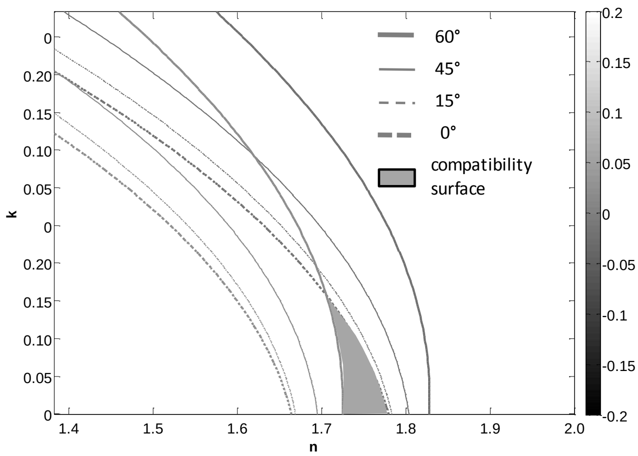

Fig. 14 Locus of refractive index giving the transmittance measured for 0°, 15°, 45°, 60° incidence angle.

As can be seen a very narrow region is now identified which gives all *n,k* couples compatible with all measured transmittances. In principle this procedure should give the “actual” refractive index (if measurements were not affected by uncertainty two angles of incidence would be enough to univocally define *n* and *k*).

For comparison purposes, the wavelength dependent real part of the refractive index, gained with the present method, is compared in Fig.15 with the profiles gained on the same sample by means of an inverse procedure based on ellipsometric measurements. Ellipsometry is usually considered very reliable and its results are often assumed as reference. The couples n,k given by the inverse procedure were exploited to evaluate the transmittance profiles of the assigned structure by means of Sentaurus simulations.

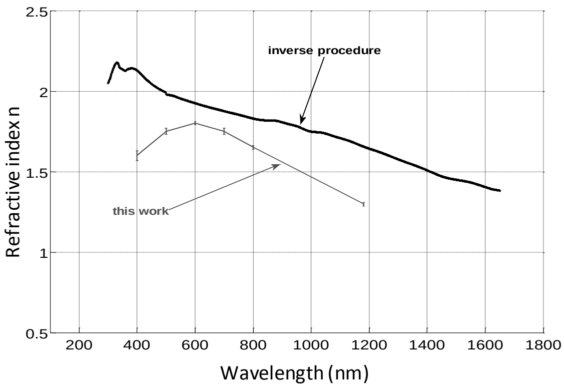

Fig. 15 Comparison between the real part of the refractive index for a ZnO on glass thin layer measured by means of an ellipsometric method (inverse procedure) and the one achieved in this work

Results are shown in Fig.16.

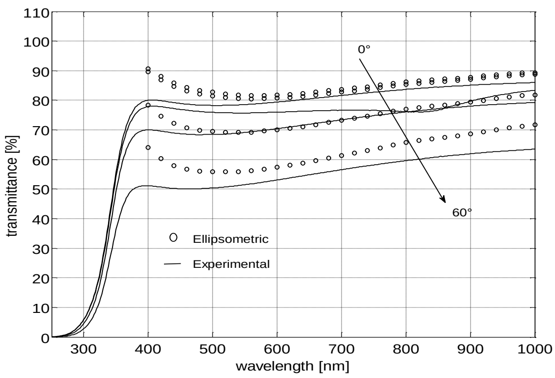

Fig. 16 Comparison between measured transmittance profiles and the transmittance profiles evaluated by Sentaurus when the refractive indexe measured by means of the ellipsometric procedure were used.

As can be seen, experiments significantly differ from numerical results, especially in the low wavelength range. On the other hand, it is worth noting that measured transmittances are perfectly coincident with those achieved by exploiting for numerical simulations *n,k* couples gained by means of the method presented in this work. This result could be considered quite trivial because we extract the couples *n,k* directly from those measurements, actually it allows a more reliable modeling of the operation of an optoelectronic device, where the main issue is the correct estimation of the wavelength dependent light availability.

5. CONCLUSIONS

In this paper an automated procedure for analyzing AFM images in the MATLAB environment has been presented. The procedure evaluates statistical parameters which qualifies the roughness of the surface and looks for the minimal area holding same parameters with the aim to simplify Sentaurus three-dimensional simulations. The procedure has allowed the extraction of an effective refractive index for ZnO thin layers. Experiments showing the overall reliability of the procedure have been shown.

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