Complete Optical Characterization of Non-Uniform SiO$_x$ Thin Films Using Imaging Spectroscopic Reflectometry

Miloslav Ohlídal
Institute of Physical Engineering, Faculty of Mechanical Engineering, Brno University of Technology, Technická 2, 616 69 Brno, Czech Republic,

Ivan Ohlídal and David Nečas
Department of Physical Electronics, Faculty of Science, Masaryk University, Kotlářská 2, Brno Czech Republic,

Petr Klapetek
Czech Metrology Institute, Okružní 31, 638 00 Brno, Czech Republic,

(Received 16 June 2008; Accepted 7 October 2008; Published 4 April 2009)

Complete optical characterization of SiO$_x$ films non-uniform in thickness is performed using imaging spectroscopic reflectometry. It is shown that by using this technique it is possible to determine the area distribution of the local thickness (area map) of these films with arbitrary shape of this thickness non-uniformity. Furthermore, it is shown that the SiO$_x$ films studied do not exhibit the area non-uniformity in dispersion (material) parameters and optical constants. This is possible because imaging spectroscopic reflectometry enables us to determine the area distributions of local thickness and local refractive index simultaneously in an independent way under the assumption that a suitable dispersion model of the refractive index of the films is used. In this paper the dispersion model corresponding to the Cauchy’s formula is used. On the basis of this dispersion model the spectral dependence of the refractive index of the SiO$_x$ films is determined. The method presented can be used to characterize the non-uniform films consisting of other non-absorbing materials. [DOI: 10.1380/ejssnt.2009.409]

Keywords: Coatings; Reflection spectroscopy

I. INTRODUCTION

A majority of thin films occurring in practice exhibits a certain degree of thickness non-uniformity along surfaces of their substrates. This is also the case of thin films prepared by various plasma chemical technologies. Thin films of SiO$_x$ prepared by plasma enhanced chemical vapor deposition (PECVD) at atmospheric pressure mostly exhibit strong thickness non-uniformity. These films are frequently employed in practice. For example, they are employed for passivation of silicon solar cells (back sides of the solar cells are covered with these SiO$_x$ films). It is known that the neglecting of thickness non-uniformity at the optical characterization of the thin films misrepresents the results of this characterization. Therefore it is important to consider thickness non-uniformity of the films in their optical characterization. So far several papers dealing with the optical characterization of various thin films exhibiting special wedge-shape non-uniformity in thickness have been published (see e.g. Refs. [1–3]). In these papers the optical characterization of the non-uniform films is based on interpretation of standard spectrophotometric data, i.e. the data corresponding to spectral dependences of reflectance or transmittance measured by standard spectrophotometers. However, in the case of the general thickness non-uniformity, i.e. the thickness non-uniformity differing from the wedge-shape non-uniformity, the procedures of the optical characterization presented in the papers cited are not usable. This means that other methods must be used to carry out the optical characterization of the thin films exhibiting this general non-uniformity in thickness. From our previous studies it followed that the methods based on utilizing imaging spectrophotometry were promising for this purpose. We developed a method employing the interpretation of the experimental data obtained using an imaging spectrophotometer working in the reflectance mode [4, 5]. This method enables us to determine simultaneously the area distributions of the thickness and optical constants (i.e. refractive index and extinction coefficient). The method is usable for determining these distributions even when general shapes of these non-uniformities occur in thin films. Thus, the method of imaging spectroscopic reflectometry (ISR) used in our earlier studies is very suitable for the complete optical characterization of the SiO$_x$ films prepared by PECVD exhibiting evident area non-uniformity. The reason of this fact is that the thickness non-uniformity of these SiO$_x$ differs from the wedge-shaped non-uniformity in general. Moreover, in principle the non-uniformity in refractive index of the thin films can be determined with the thickness non-uniformity simultaneously when ISR is used. In this paper results concerning the complete optical characterization of SiO$_x$ film serving for passivation of silicon solar cells will be presented.

II. EXPERIMENTAL

The SiO$_x$ thin films were prepared by PECVD on silicon single crystal substrates. Within PECVD a linearly extended DC arc discharge and microwave plasma source were used to activate the processing gases. An Ar-N$_2$ mixture was fed through the plasma source and precursors were injected into the activated plasma gases outside...
the plasma source nearby the substrate. Controlled purge gas systems prevented a contamination of a reaction zone with air or moisture as well as the release of reaction products. The concrete deposition parameters employed were as follows. Plasma: 24 slm Ar/10.5 slm N₂, precursor: TEOS, substrate heating in temperature of 200°C.

For measuring the experimental data the self-made two-channel spectroscopic imaging reflectometer containing a CCD camera as a detector was employed. The reflectance experimental data was measured at normal incidence of light within the spectral region 280-730 nm. A detailed description of this arrangement is presented in our earlier paper [5].

III. DATA PROCESSING

Owing to the fact that the dimensions of the local areas on the non-uniform film corresponding to individual pixels of the CCD camera are relatively very small, it is possible to assume that over these areas the film is uniform. This means that the spectral dependences of the reflectance $R_{k,j}^{s}$ measured by a pixel are given by the formulae valid for the uniform films ($k$ and/or $j$ denotes the $k$ th row and/or $j$ th column in which a certain pixel is placed in the matrix of the CCD chip), i.e. [6, 7]

$$R_{k,j}^{s} = |r_{k,j}^{s}|^2,$$

where

$$r_{k,j}^{s} = \frac{r_{k,j}^{s} + r_{k,j}^{s} \exp(i\pi_{k,j})}{1 + r_{k,j}^{s} r_{k,j}^{s} \exp(i\pi_{k,j})},$$

$$r_{k,j}^{s} = \frac{n_0 - n_{k,j}^{s}}{n_0 + n_{k,j}^{s}}, \quad r_{k,j}^{s} = \frac{n_{k,j}^{s} - n_{k,j}^{s}}{n_{k,j}^{s} + n_{k,j}^{s}}, \quad x_{k,j}^{s} = \frac{4\pi n_{k,j}^{s} d_{k,j}^{s}}{\lambda}. \quad (3)$$

In the foregoing equations symbols $n_0$, $\bar{n}$, $n_{k,j}^{s}$, $d_{k,j}^{s}$ and $\lambda$ denote the refractive index of the ambient, complex refractive index of the substrate, local refractive index of the film, local thickness of the film and wavelength of incident light, respectively. Note that the ambient consists of air and therefore $n_0 = 1$. The complex refractive index of the substrate is expressed as $\bar{n} = n - ik$, where $n$ and $k$ is the real refractive index and extinction coefficient of the substrate, respectively. The foregoing mathematical expression for the local non-uniform film reflectance $R_{k,j}^{s}$ belonging to the individual pixels of CCD camera corresponds to the reflectance of the non-absorbing thin film deposited onto an absorbing substrate. This is given by the fact that the SiO₂ thin films are assumed to be non-absorbing within the entire spectral region of interest, i.e. within spectral region 280-730 nm. This assumption is supported by our previous optical studies of these films.

The least square method (LSM) was used to treat the experimental data using Eq. (1). Of course, the LSM had to be applied for each local area on the film corresponding to the given pixels of the CCD camera. Within the LSM, the following merit function $S_{k,j}^{s}$ was employed:

$$S_{k,j}^{s} = \sum_{s=1}^{K} \left( \frac{R_{k,j}^{s} - R_{k,j}^{s}}{\sigma_{k,j}^{s}} \right)^2, \quad (4)$$

where $R_{k,j}^{s}$ and/or $R_{k,j}^{s}$ denotes the theoretical and/or experimental value of the local reflectance corresponding to the $(k,j)$th pixel and wavelength $\lambda_s$. Symbol $\sigma_{k,j}^{s}$ and/or $K$ represents the standard deviation of $R_{k,j}^{s}$ and/or $K$ the number of the measurements of the reflectance, i.e. the number of wavelengths, for which the reflectance was measured in each pixel.

Within the processing of the experimental data, the dispersion model corresponding to the Cauchy’s formula [8] was used to express the spectral dependences of the refractive index of the non-uniform SiO₂ films.

Thus, the local refractive index of the non-uniform SiO₂ films was expressed as follows:

$$n_{k,j}^{s} = A_{k,j}^{s} + B_{k,j}^{s} \frac{\lambda^2}{\lambda^2}, \quad (5)$$

where symbols $A_{k,j}^{s}$ and $B_{k,j}^{s}$ are the local dispersion (material) parameters of the films studied.

From the foregoing it is evident that using the LSM the values of the local thickness $d_{k,j}^{s}$ and local dispersion parameters $A_{k,j}^{s}$ and $B_{k,j}^{s}$ were determined for each local area of the SiO₂ films. Using the local dispersion parameters found one can calculate the true spectral dependences of the local refractive index of the non-uniform SiO₂ films. This means that area distributions (maps) of the thickness and refractive index of the SiO₂ films can be determined in this way. Note that the spectral dependences of the optical constants of the silicon single crystal substrates were taken from the literature [9] and fixed within LSM.

IV. RESULTS AND DISCUSSION

In this section the results of the optical characterization of the selected non-uniform SiO₂ film are presented. These results are typical of the optical characterization of these films being studied using ISR.

In Fig. 1, the CCD image of the sample of the SiO₂ film under study is plotted for the wavelength 450 nm. In this figure the dark and bright areas are seen which indicate an area optical non-uniformity of the film studied.

In Fig. 2, the local thickness distribution (map) corresponding to the selected area of the non-uniform SiO₂ film is presented. This figure shows that the selected SiO₂ film exhibits the strong thickness non-uniformity. The local thickness distribution presented in Fig. 2 corresponds
The distribution of the local thickness of the SiO<sub>x</sub> film under investigation.

The local peaks in the local thickness distribution in Fig. 2 are caused by dust particles on the film and local defects of the film. By means of the LSM values of the local dispersion parameters <i>A</i><sup>k,j</sup> and <i>B</i><sup>k,j</sup> were determined together with the local thickness <i>d</i><sup>k,j</sup>. It was found that the values of the dispersion parameters were practically identical for all the local areas belonging to individual pixels. This fact implies that the non-uniform SiO<sub>x</sub> thin film under investigation does not exhibit the area non-uniformity in dispersion parameters. From this fact it is also possible to deduce that the film studied is uniform in refractive index along the substrate area. One can thus state that the SiO<sub>x</sub> film analysed exhibits only the area non-uniformity in thickness. This conclusion could be achieved on the basis of the fact that this characterization method allows us to determine the distributions of the local thickness and local optical constants in an independent way. The values of the dispersion parameters found for all the pixels are as follows: <i>A</i><sup>k,j</sup> ≡ A = 1.208 and <i>B</i><sup>k,j</sup> ≡ B = 11537.4 nm<sup>2</sup>. The spectral dependence of the refractive index characterizing the selected SiO<sub>x</sub> film non-uniform in thickness calculated using the values of the dispersion parameters found is plotted in Fig. 3.

In Fig. 4 the spectral dependence of <i>R</i><sup>k,j</sup> of SiO<sub>2</sub> film selected corresponding to the chosen pixel is plotted. In this figure it is seen that there is a relatively good agreement between the experimental and theoretical data. The theoretical data were calculated using the values of the parameters found, i.e. using the values of <i>A</i>, <i>B</i> and <i>d</i><sup>k,j</sup> (<i>d</i><sup>k,j</sup> = 213 nm). The similar agreement between theoretical and experimental data was also observed for the remaining pixels. This agreement supports a correctness of the results of the complete optical characterization of the non-uniform SiO<sub>x</sub> selected, i.e. the correctness of the thickness distributions (maps of the local thickness) and spectral dependence of the refractive index of the non-uniform SiO<sub>x</sub> film under study.

From Fig. 3 it is seen that the values of the refractive index of the SiO<sub>x</sub> film investigated are lower than expected. This fact can be caused by several reasons. We mean that the most probable reason consists in the existence of the columnar structure of the SiO<sub>x</sub> films studied. In this case there are pores among the columns consisting of the material of the film. This means that the refractive index determined (see Fig. 3) corresponds to the “effective” (mean) values of this quantity. The values of the refractive index of the columns and pores (pores can be partially filled with water or other liquids) together with the packing density characterizing the relative total volume of the columns in the film cannot be determined using ISR in principle. These quantities can only be determined using an application of ellipsometric or photometric porosimetry. If we want to perform a very rough estimation of the packing density of the SiO<sub>x</sub> film studied we will assume that the columns are formed by amorphous SiO<sub>2</sub> and pores are filled with air. Then the following formula for the “effective” (mean) refractive index <i>n</i><sub>1</sub> of the columnar film can be used:

\[ n_1 = pn_{1s} + (1 - p)n_0, \]  

where symbols <i>p</i> and <i>n</i><sub>1s</sub> denote the packing density and columnar refractive index, respectively. For our estima-
tion it is put that \( n_0 = 1 \), \( n_1 \) is identical with the literature data corresponding to amorphous SiO\(_2\) and \( n_1 \) is identical with the values determined (see Fig. 3). Then we obtain: \( p \approx 0.7 \) (porosity \( 1 - p \approx 0.3 \)).

From the foregoing results it is seen that ISR is the efficient method for the complete optical characterization of the SiO\(_x\) films exhibiting area non-uniformity in thickness. If the SiO\(_x\) films exhibit area non-uniformity in both thickness and refractive index simultaneously, the method presented will be successful too. Of course, this method will also be efficient and successful for other non-uniform thin films consisting of non-absorbing materials.

V. CONCLUSIONS

This paper presents the results concerning the complete optical characterization of non-uniform SiO\(_x\) films prepared by atmospheric pressure PECVD onto the silicon single crystal substrates obtained by means of ISR. The results presented in this paper concern the selected sample of the non-uniform SiO\(_x\) film. The similar results were obtained for other samples of these films. From the results of the optical characterization it is concluded that these SiO\(_x\) films exhibited area non-uniformity in thickness only. It is shown that the SiO\(_x\) films under study are uniform in the refractive index along the areas of their substrates. The distribution of the local thickness (map of the local thickness) is presented for the selected sample for illustration. The spectral dependence of the refractive index of this sample calculated using the values of the dispersion parameters found is presented too. Moreover, it was shown that the values of the local thickness and values of the dispersion parameters could be found using the LSM simultaneously and independently for all the local areas corresponding to the individual pixels of the CCD camera. Thus, the area distributions (maps) of the local thickness and local refractive index can be found simultaneously and independently in general. The same conclusion is valid for the non-uniform non-absorbing thin films consisting of other materials. Thus, it is possible to claim that ISR is the useful method for the optical characterization of the non-absorbing thin films exhibiting area non-uniformity.

Acknowledgments

This work was supported by the Ministry of Education, Youths and Sports of the Czech Republic under contracts MSM 0021630518, MSM 0021622411 and the Ministry of Industry and Trade of the Czech Republic under contract FT-TA3/142. We thank to A. Poruba and R. Barinka for providing us the samples prepared within the European project FP6 COOP-CT-2005-017586.

[1] M. I. Török, Opt. Acta 32, 479 (1985).
[2] T. Pisarkiewicz, T. Stapinski, H. Czternasek, and P. Rava, J. Non-Cryst. Solids 137/138, 619 (1991).
[3] T. Pisarkiewicz, J. Phys. D: Appl. Phys. 27, 160 (1994).
[4] M. Ohlídal, I. Ohlídal, P. Klapetek, M. Jákl, and V. Cudek, Jpn. J. Appl. Phys. 42, 4760 (2003).
[5] M. Ohlídal, V. Cudek, I. Ohlídal, and P. Klapetek, in Advances in Optical Thin Films II, Vol. 5963 SPIE, Eds. C. Amra, N. Kaiser and H. A. Macleod (Bellingham, Washington, 2005) p. 596329-1.
[6] A. Vašíček, Optics of Thin Films, 2nd ed. (North Holland, Amsterdam, 1960).
[7] Z. Knittl, Optics of Thin Films, 1st ed. (John Wiley, Chichester 1976).
[8] M. Born and E. Wolf, Principles of Optics, 7th ed. (Cambridge University Press, Cambridge, 2005).
[9] C. M. Herzinger, B. Johns, W. A. McGahan, J. A. Woolan, and W. Paulson, J. Appl. Phys. 83, 3323 (1998).
[10] H. R. Phillip, in Handbook of Optical Constants of Solids, Ed. E.D. Palik (Academic Press, San Diego, 1999) p. 749.