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Ellipsometric characterization of SiO_x films with embedded Si nanoparticles

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A B S T R A C T

In this work we present results on the ellipsometric study of SiO_x films in the spectral range of 280–820 nm. The films were deposited by vacuum thermal evaporation of SiO onto Si substrates heated at 150 °C. To stimulate the formation of silicon clusters in the oxide matrix the films were annealed at temperatures 700, 1000 and 1100 °C in argon for 5, 15 and 30 min. By applying the Bruggeman effective-medium approximation theory and using multiple-layer optical models, from the ellipsometric data analysis the thickness, complex refractive index and composition of the films, as well as the size of the embedded Si nanocrystallites have been determined. Atomic-force microscopy imaging showed a very smooth surface, the roughness value of which correlated well with the top-layer thickness, determined from the ellipsometric data analysis.

1. Introduction

Nano-sized Si clusters have received a great deal of attention because of their wide area of application. Nowadays, it is well-established that using Si nanocrystals (nc-Si) in optoelectronic and microelectronic applications is more and more desirable. In recent years the properties of nanocrystalline Si thin films have been extensively studied due to the observed intensive visible-light emission at room temperature. Appearance of visible photoluminescence (PL), related to nc-Si clusters embedded in an amorphous SiO_x, matrix opens a new area of application of SiO and SiO_2 films. Nanostructured SiO_x films exhibit a number of advantages, such as high stability and contemporary with complementary microelectronic technology.

The process of formation of SiO_x films with embedded Si nanoparticles generally consists of two stages: (i) production of SiO_x films by different techniques, such as thermal [1–4], electron gun evaporation of silicon monoxide (SiO) [5] or implanting Si atoms in SiO_2 layers [6–8]; (ii) annealing of the films to promote formation of nanometer-sized Si clusters. The annealing temperature determines the structure of Si inclusions. Annealing at 500–900 °C favours the coagulation of Si atoms into amorphous clusters. At higher temperatures the Si inclusions start to crystallize, as the size of the Si nanocrystallites depends on the annealing conditions [3,4]. Therefore, control of these parameters is of great importance for application of silicon oxide films in light emitting Si-based devices.

Spectroscopic ellipsometry (SE) is a powerful nondestructive tool for detection of thermally stimulated changes in SiO_x structure and composition. Correlation between the optical constants and film structure can be established by analyzing the complex dielectric function ε of nanostructured SiO_x films, since ε strongly correlates with the energy band structure of the film. Hence, any change in the film microstructure results in modification of the shape and position of dielectric function features, associated with electronic transitions that contain information on film structure and, therefore, additional information about the formation of nano-sized Si inclusions can be derived. Recently, much effort has been made to determine the real dielectric function of nc-Si, which depends strongly on crystallite-size [5,9–12]. It has been shown that a decrease in the nanocrystallite size causes a shift of the dielectric function towards higher energies [5,9–12]. Calculations have been made by fitting the SE measurements data with an appropriate model. The most frequently used models are based on the Bruggeman effective-medium approximation (BEMA) theory [5,9,10,12,13], by which a homogeneous medium can be well described.

In this paper we have applied spectroscopic ellipsometry to study evaporated SiO_x films in order to gain information about the formation of Si nanoparticles in the oxide matrix. The ellipsometric

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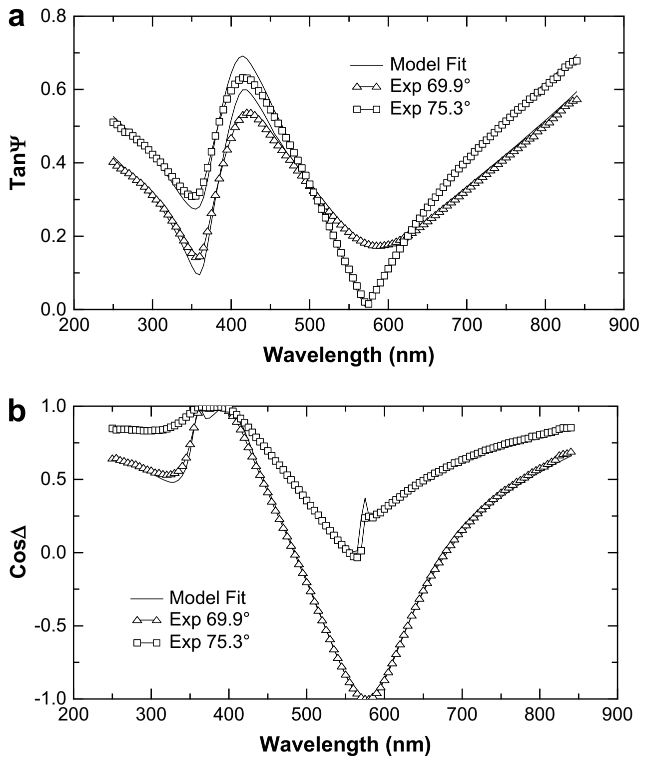

Fig. 1. Experimental and theoretical data of tanΨ (a) and cosΔ (b) functions at two angles of incidence, inserted. The SE measurements were done on 164 nm thick SiO_x film, annealed at 1000 °C for 5 min.

data, however, is not meaningful by itself and, in order to get useful information about the film composition and size of Si nanocrystallites, the films are examined with multiple-layer optical models by applying the BEMA theory. To support the validity of the optical models in this study we performed atomic-force microscopy (AFM) measurements and the correlation of the results of the SE data analysis are discussed.

2. Experimental details

Silicon monoxide powder (Cerac Inc., purity of 99.9%) was evaporated at a residual pressure of $\sim 10^{-3}$ Pa on monocrystalline silicon (c-Si) substrates kept at 150 °C. In order to stimulate the formation of Si nanoparticles in the evaporated SiO_x, the films were thermally annealed in an Ar atmosphere at 700, 1000 and 1100 °C for 5, 15 and 30 min.

After each technological step, the ellipsometric measurements were carried out on a “Rudolf Research” variable-angle manual ellipsometer in the wavelength region of 280–820 nm and at different angles varying from 50 to 75°. The accuracy of the incident angle and the polarization angles is 0.01°. By fitting the measured Ψ and Δ angles data with an appropriate model using the BEMA theory, the thickness, optical constants and the physical composition of the films were determined. The film was considered as consisting of several sub-layers, each having different thickness and composition. The calculation procedure involved the selection of the reasonable starting thickness and composition of each sub-layer and introduction of reference dielectric functions of the chosen components, taken from the literature. Each sub-layer was considered as a physical mixture of components, such as SiO [14], SiO_2 [14], and either a-Si [14], c-Si [15] or fine-grain poly-Si [16] and voids in some degree. The evaluation software allowed the simultaneous search for the thickness and composition of the sub-layers. An iterative least-squares method was used for minimizing the difference between the experimental ellipsometric angles Ψ and Δ and the theoretical ones. As the correct optical model was selected and tested by comparing the measured and calculated spectral dependence of Ψ and Δ angles, the best fit resulted in a minimum mean square error (MSE).

Table 1
Film composition and volume fraction of the components.

Annealing conditions	Volume fraction of the components (%)					Oxide state
	f_{SiO}	f_{SiO_2}	$f_{a\text{-}Si}$	$f_{poly\text{-}Si}$	f_{voids}	
[a]before annealing	74.0	22.2	0.2	–	3.6	$SiO_{1.12}$
[a]700 °C, 5 min	75.8	22.4	1.8	–	–	$SiO_{1.23}$
[b]700 °C, 30 min	71.7	24.0	4.3	–	–	$SiO_{1.26}$
[b]1000 °C, 5 min	53.3	33.5	–	13.2	–	$SiO_{1.38}$
[b]1000 °C, 30 min	33.1	52.7	–	14.2	–	$SiO_{1.61}$
[b]1100 °C, 15 min	–	77.7	–	22.3	–	SiO_2

[a] Single-layer optical model.
[b] Two-layer optical model.

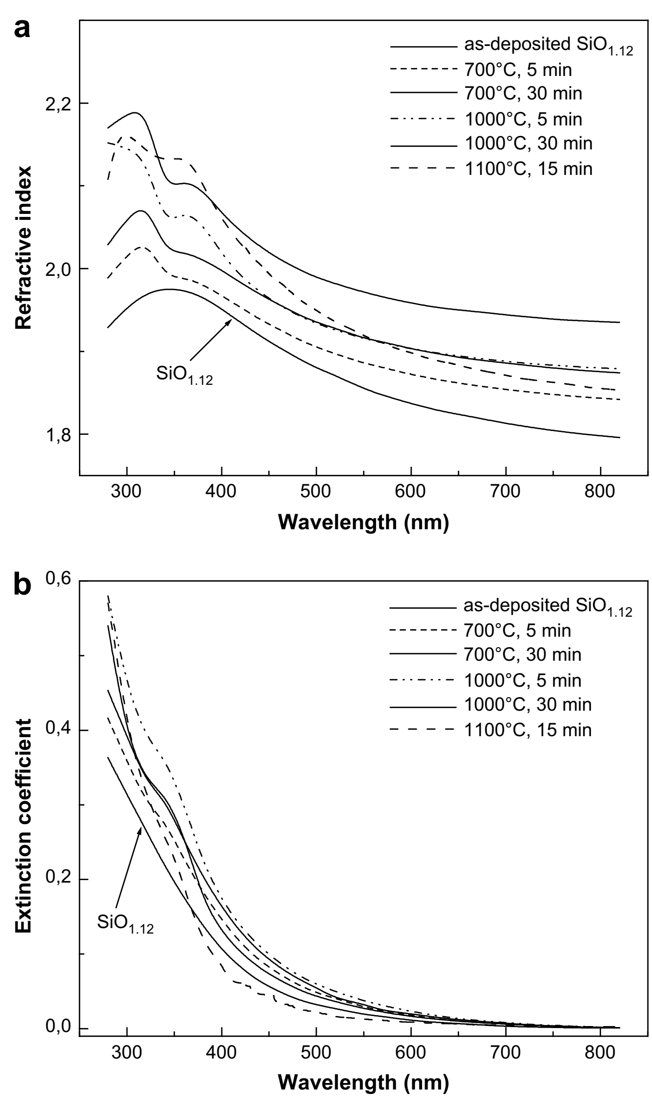

Fig. 2. Dispersion functions of the refractive index (a) and the extinction coefficient (b) of SiO_x films before and after annealing.