Ellipsometric studies of thin silver films deposited by DC magnetron sputtering[1]

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Abstract – The structure and the growth mechanism of silver films of the thickness up to 20 nm deposited onto glass substrates by DC magnetron sputtering were examined in the paper. Film optical properties were investigated using spectroscopic ellipsometry in the wavelength range of 400–800 nm. A three-layer model allowing determining the average thickness and the structure of thin silver films has been developed. The data obtained by means of this model have good agreement with results of measurements carried out using an atomic force microscope. Both methods have shown that silver films on glass grow by Stranski-Krastanov mechanism.

1. Introduction

Study of the structure and properties of ultrathin films acquire increasing importance since such films have unique physical properties and are used in different electronic, magnetic, and optical devices [1, 2].

There are a number of publications devoted to study of growth mechanism of silver films deposited by different methods [3–6]. However, in most of them attention is given to coatings at semiconductor substrates. The following methods were used to investigate the films: low energy electron diffraction, scanning tunneling microscopy [7], transmission electron microscopy [8], atomic force microscopy [9], x-ray reflectivity [10], etc.

The structure and the growth mechanism of thin silver films at glass substrates were investigated in the present work. Film deposition was made by DC magnetron sputtering at a room temperature. The main research methods were spectroscopic ellipsometry and atomic force microscopy.

2. Experimental

Film deposition was produced by a magnetron sputtering system with a silver cathode of 120-mm diameter. A vacuum chamber was preliminary pumped out using a turbo-molecular pump to the residual pressure of 10^{-3} Pa. Immediately before the process of film deposition the substrates were subjected to the ion-plasma cleaning made by means of the ion source with the closed electron drift. Film deposition was realized at the argon pressure of 0.09 Pa. The discharge voltage and the current were equal to 400 V and 1.3 A, respectively. The magnetron-substrate distance was 12 cm. Two films denoted further as Film 1 and Film 2 were deposited at equal conditions with deposition times of 6 and 12 seconds, respectively.

Information concerning surface morphology of the deposited films was obtained using the atomic force microscope Solver P47. A probe with a point having a curvature radius less than 10 nm was used in the measurements. Resulting from the measurements, the film thickness as well as the shape, dimension and distribution of silver islands over the substrate surface were determined.

Main attention was given to investigation of optical characteristics of silver films by means of a fast spectroscopic ellipsometer in the wavelength range of 400–800 nm. The ellipsometry method principle is in measurement of change of the state of light polarization after its reflection from the sample surface. The results of measurement are the ellipsometric angles Ψ and Δ functionally related to the optical parameters (refraction and absorption indices, film thickness) determined from these angles by means of mathematical calculations.

3. Results

According to the reference data, silver films have two main growth mechanisms depending on their thickness and deposition conditions [11]. The first is the island mechanism by Volmer-Weber characterized by beginning of the film growth with appearance of separated three-dimensional nuclei that later are joined into a continuous layer. Another mechanism called layer plus island was suggested by Stranski-Krastanov. Its main distinction is that at the initial moment of time the film grows layer-by-layer but then this growth is disturbed and the film keeps on growing by the island mechanism.

Island mechanism of silver film growth was observed at the deposition of such films onto the Ge(001) [3], GaAs(110) [4], Si(001) [10] substrates. The layer plus island mechanism took place in case

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of the silver film deposition onto Si (111) substrates [12, 13].

Investigation of thin silver films by means of spectroscopic ellipsometry has shown that it helps to determine the film growth mechanism at the initial stage of their growth.

Ellipsometric investigations consisted from three successive stages including obtaining of experimental data, structural simulation and obtaining of model-based calculation data, change of the model parameters for the best coincidence of experimental and calculation results. The following model parameters were changed: angle of incidence of the beam of light, quantity of layers, their thickness and percentage of materials in them.

Values of ellipsometric angles Ψ and Δ as a function of the incident light wavelength were received resulting from the ellipsometric measurements (Figs. 1 and 2). The angle of incidence of the beam of light was equal to 70°. The diagrams present the experimental results (open symbols) and the calculated data (filled symbols). The samples with Film 1 and Film 2 as well as the 1500-nm thick silver film were subjected to investigation. The letter film was sufficiently thick to have the properties of a bulk material. The obtained values of Ψ and Δ for a thick film have good agreement with the data presented in Ref. [14]. It is seen from the diagrams that with the film thickness increase the values of ellipsometric angles typical for this film approach the values inherent to a bulk material.

Calculated values of the angles Ψ and Δ were received on the basis of a three-layer model presented in Fig. 3. As it is seen from the Figs. 1 and 2, this model allow to obtain good agreement of calculated and experimental data. The model includes one glass layer simulating a substrate and two silver layers presenting a film. These two silver layers consist of a thin continuous layer and a thicker layer with gradient content of void. The gradient layer was used to simulate the islands situated at a continuous film layer. Void percentage in the gradient layer was the least at the layer base and the highest at its upper part. Thus, island cross-section was presented in the form of a trapezium (fig. 3). The ratio of the upper and lower sides of this trapezium was determined by the void content in the film.

According to the suggested model, the investigated films had the following structure:

Film 1 – a continuous layer (1.5-nm thick) + a gradient layer (14-nm thick) containing 46% of voids at the low boundary and 14 % at the upper one.

Film 2 – a continuous layer (3-nm thick) + a gradient layer (14-nm thick), containing 66% of voids at the low boundary and 32 % at the upper one.

Necessity of a thin continuous layer in the model allows supposing that silver films on glass begin growing by Stranski-Krastanov mechanism.

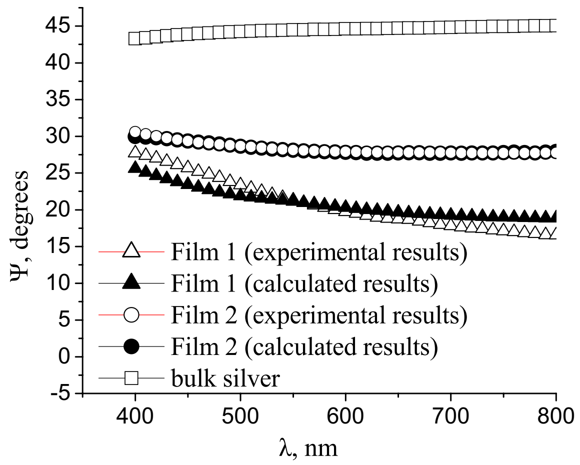


Fig. 1. Measured and calculated values of the ellipsometrical angle Ψ as a function of light wavelength λ

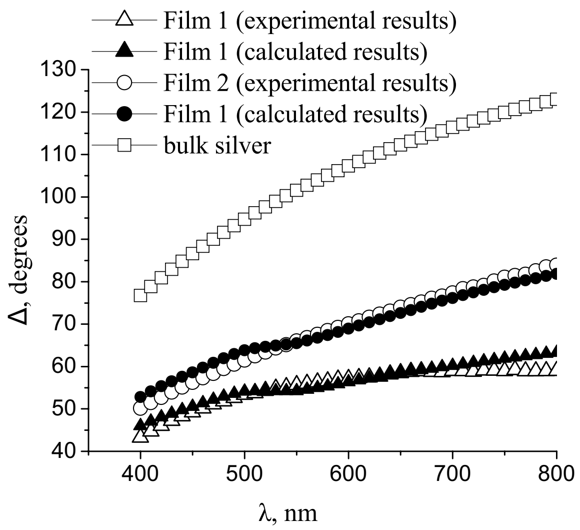


Fig. 2. Measured and calculated values of the ellipsometrical angle Δ as a function of light wavelength λ

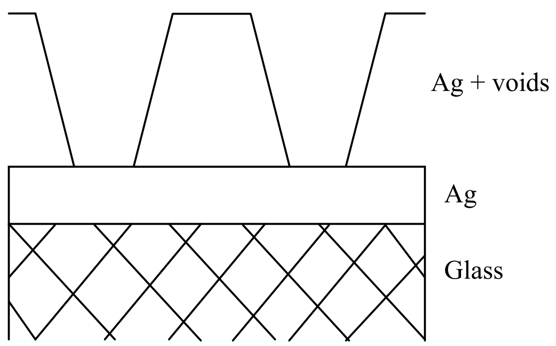
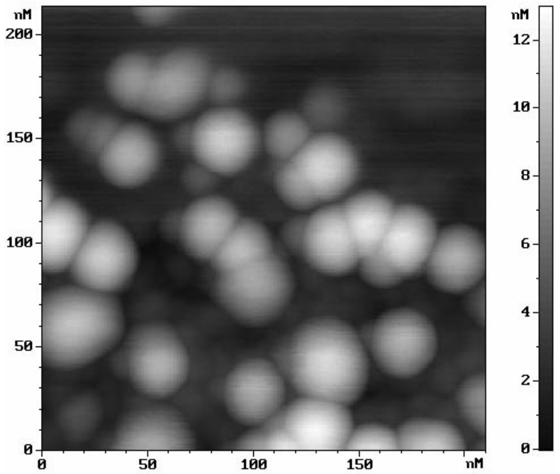


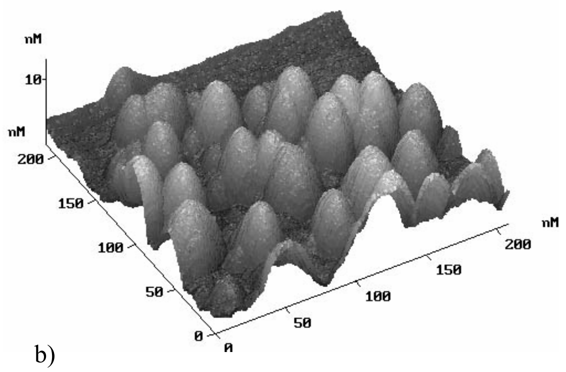
Fig. 3. Three-layer model for analysis of thin silver films

In due course, the transverse growth of islands is observed, their height being kept the same. The latter phenomenon was already described in references [13].

To check the accuracy of the three-layer model used at ellipsometric measurements, silver films were investigated by means of the atomic force microscope. Fig. 5 presents the images of the Film 2 surface. It was discovered that the film consists of a continuous layer of approximately 8-nm thickness at which the islands of 20–30-nm diameter and 6-nm height are disposed. These results confirm the layer plus island mechanism of the film growth.



a)



b)

Fig. 4. AFM image of Film 2 surface. a) top view, b) 3D image

In spite of the fact that some lack of coincidence in the thicknesses of the continuous and island parts of the film is observed, the results obtained by means of two principally different methods testify to new possibilities of spectroscopic ellipsometry application for study of the growth mechanisms of island silver films.

Conclusion

Investigation of silver films of the thickness up to 20 nm deposited onto the glass substrates by the DC magnetron sputtering method was carried out in the given work by means of spectral ellipsometry. It is shown that the film growth begins from formation of a continuous silver layer of the thickness of several nm. Then, resulting from the stresses generated in this layer, layer-by-layer film growth is disturbed and formation of the half-sphere shape islands begins. This film growth mechanism was testified both by the three-layer model used at ellipsometric measurements and by the film surface images obtained by means of the atomic force microscope.

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