Change Spectrum Characteristics Modification of Films Deposited by Magnetron Sputtering with the Assistance of Argon Ions Beam

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Abstract. Thin aluminum films were prepared using the method of magnetron sputtering with and without argon ion beam assistance. The influence of argon ion beam on the reflectivity in the UV range and the structure of aluminum films was studied. The structure of the films was studied by transmission electron microscopy (TEM), X-ray diffractometry (XRD) and atomic-force microscope (AFM). The study has shown that the films deposed with the assistance of the argon ion beam have more significant microstresses associated with an increase of crystallites microstructure defects as compared to the films deposed without ion assistance. Comparison of the measured reflectivity of aluminum films deposed without and with the assistance of the ion beam has shown that the films characterized by a higher level of microstructure def ects have increased reflectivity in the UV range. The studies suggest that the defects of thin aluminum films crystal structure influence its optical properties.

1. Introduction

High-reflectance Al thin films have been widely used in optics and optical applications. Optical characteristics of these coatings have been studied quite extensively. The effects of such parameters as film thickness and deposition conditions (including pressure, deposition rate, substrate temperature, and residual gasses) have been previously examined in several papers [1-4]. Additionally, as described in the studies [5, 8], optical properties of the films can vary depending on the deposition method (magnetron sputtering, electron-beam evaporation, thermal deposition, laser deposition, etc.), which is connected with the modification of their structure. In turn, the structure of a film coating is highly determined by the initial stage of the crystallites' nuclei formation and growth processes [8, 9]. Upon formation the crystallite structure may change as a result of plastic deformation, exposure to neutral or charged particle radiation, or electromagnetic radiation. Thus, the application of an assist ion beam when depositing film coatings leads to their structure modification, which in turn results in the modification of their optical properties [6, 7, 10,11].

The results obtained in the above mentioned studies demonstrate the potential of using charged particle beams when depositing film coatings in order to modify their structure with a view to

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changing their optical characteristics. It is also worth noting that the relationship between the structural changes and optical parameters has been relatively understudied due to the variety of the accompanying processes.

This paper investigates the effect of the concomitant Ar ion irradiation of the Al films deposited by magnetron sputtering on the modification of their reflectance in the UV region.

The relationship between the reflectance and the crystalline structure is not always obvious. Structure modification induced by any reason results in the modification of the solid-state plasma parameters. These modifications change the nature of the electromagnetic field interaction with matter. In our study, the films' reflectance was measured by a photometer. The TEM, XRD, and AFM methods were used to examine the Al films' structure and topology. The reflection factors of the Al films produced with and without Ar ion-beam assist were compared.

2. Experimental details

Al thin films were deposited by the magnetron sputtering process with a flat target made of high-purity aluminium (99.99 wt.%). The films were deposited on glass substrates previously cleaned with a chalk suspension in isopropyl alcohol. Argon with 99.996 wt.% was used as the sputtering gas. The gas flow was controlled by an electronic flow meter; the gas pressure was measured by a thermocouple-ionization vacuum gage. Prior to the deposition process, the aluminium target was cleaned by scattering during 4 min. All the samples were prepared at a pressure in the vacuum chamber of $8x10^{-4}$ Pa before sputtering and $1.4x10^{-1}$ Pa during sputtering.

On one part of the samples the Al films were deposited by using only magnetron sputtering while on the other part of the samples the films were deposited with the use of the Ar ion-beam assist at the initial stage of the deposition process. The ion beam was perpendicular to the sample surface. The energy characteristics of the beam are given in Figure. 1.

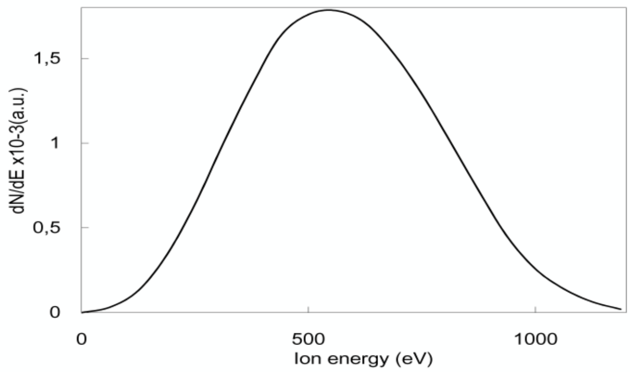

Figure 1. Ar ions distribution in the beam with regard to the energies

Upon assisting the ion current was 320 mA. In both cases the thickness of the deposited Al films was approximately 85 nm; the deposition rate was 7.5 nm/s.

The reflectance spectra of the grown films were measured by the SF-256 spectrophotometer (LOMO, Russia) with the use of a specular reflectance attachment. Five samples of each type were prepared for the investigation; each sample was prepared in a separate sputtering process under similar deposition conditions.

The TEM, XRD and AFM measurements were carried out by using the CM 12 Transmission Electron Microscope (PHILIPS, Netherlands), XRD6000 X-ray Diffractometer (SHIMADZU, Japan), and Solver HV Atomic Force Microscope (NT-MDT, Russia) correspondently.

3. Results and discussion

The samples were held in air for more than 120 hours. Then, the Al films optical characteristics were measured. The results of the Al films' reflection factor measurements in the 200-240 nm spectrum range for the samples obtained with and without ion assist are given in Figure. 2. It is seen that the reflection factor of the Al films produced by ion-assisted deposition is 4 – 5% higher than that of the films obtained without ion assist.

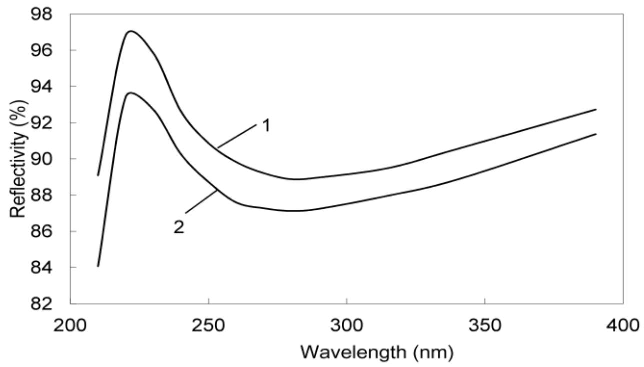

Figure 2. Al films reflectivity: 1 – with assistance; 2 – without assistance

The reflection factor of a coating is known to depend on a number of factors: topography, crystallite size, and the level of the crystal lattice perfection. To determine the reason that accounted for the increase in the reflection factor we performed the TEM, AFM, and XDR analyses of the obtained films.

The conducted TEM analysis showed that the Al films had similar nanocrystallite structure. The size of the Al crystallites was 15 – 470 nm for the films deposited without ion assist and 70 – 300 nm for the films deposited with ion assist.

In the case of ion assist, the change in the size of the crystallites at upper and lower borders is caused by the impact of the Ar ions on the processes that occur upon the condensation of aluminium on the deposited surface area. Under the impact of an ion beam the formed islands get partially or fully sputtered on the deposited surface, which leads to an increase in the adsorbed aluminium atoms' concentration. This process is schematically shown in Figure. 3.

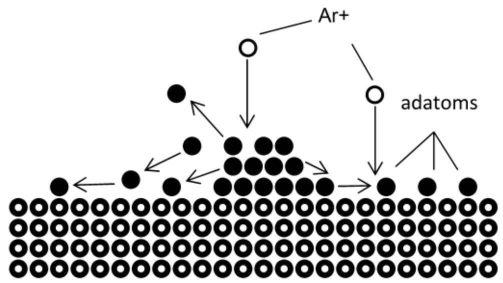

Figure 3. Scheme of Ar ion interaction with the surface

Large islands are sputtered only partially while s mall islands of a critical size get fully sputtered. It is also worth noting that the ion-assisted processes are influenced not only by the ions' energy but also by the density of the ion current [7]. The additional formation of the adsorbed atoms contributes to the growth of the small, unsputtered islands. Thus, as a result of the competing processes, islands with a less size spread are formed on the surface. The result of the ion assist application is the increased crystallite size at the lower border and reduced crystallite size at the upper border. It coincides with the experimental results described in [7].

To determine the microrelief of the deposited film surface we have performed the AFM studies. Figure.4 shows the AFM images of the Al films deposited with and without ion assist. As seen in this figure, the surface roughness of the films produced by the ion-assisted deposition slightly increases. Indeed, the root-mean-square surface roughness of the films deposited without ion assist is 2.289 nm, with ion assist – 2.397 nm.

The conducted TEM and AFM examination of the Al films produced by ion-assisted deposition has shown that the resultant changes in the crystallite size and roughness are insignificant. Hence, the geometrical factor cannot be the reason for the reflectance growth.

Another reason for the reflectance modification can be the imperfection level of the crystalline structure since real crystals are always characterized by the presence of defects. The defect concentration depends on the conditions of the crystalline structure formation. Under the impact

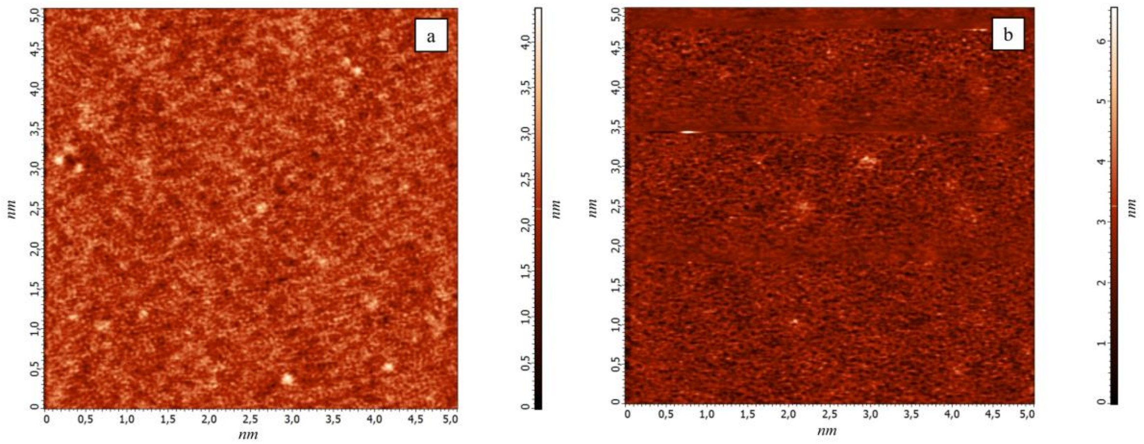

Figure 4. AFM images: 1 – without assistance; 2 – with assistance

of the assist ions of a relatively high energy, point defects (vacancies and dislocated atoms) are formed in the growing film. The defects of given type have a considerable effect almost on all the properties of solid bodies including optical characteristics. Point defects in crystals provide a slight lattice distortion, which, in turn, results in the appearance of internal microstress. The microstress value is known to be proportional to the density of the defects. Consequently, the density of the defects in a film can be estimated by the microstress value.

We have conducted the XRD studies of the films obtained by various deposition methods. The diffraction patterns have been analysed by using the Williamson-Hall method. The measurement results are given in Figure 5 and Table 1. As seen from the table, the microstress value is more than 5 times higher in the films produced by the ion-assisted deposition. Hence, there is an increase in the concentration of the crystalline structure defects of the film coating.

Table 1. XDR measurement results

	scaling lattice, a, $x10^{-10}$, m	coherent scattering region (CSR), x 10^{-9} m	relative strain, x 10^{-3}	preferred orientation, March-Dollase (111)
without assistance	4.0550	15	1.5	0.5350
with assistance	4.0161	62	8.5	0.7517

A point defect interacts with all the conduction electrons. The electrons located in the defect's neighborhood are "bound" by it and do not take part in conductivity, i.e. the N_e electron concentration decreases. This leads to the decrease in the plasma frequency ω_p, electron collision rate Y, the parameter $Q = v_e w_p /c$ that characterize the influence on the absorption of the spatial dispersion effects of the conductivity, etc. Thus, the parameters of the solid-state plasma undergo changes. Since the reflection factor R(w) and the absorption factor A(w) are determined by these parameters, their modification leads to the change in the nature of the electromagnetic wave interaction with matter. In the UV region, the role of the forced oscillations of the crystal lattice bound electrons that intensely absorb light in the area close to the resonance frequency is increased. The reflection and absorption factors in this area are highly dependent on the frequency.

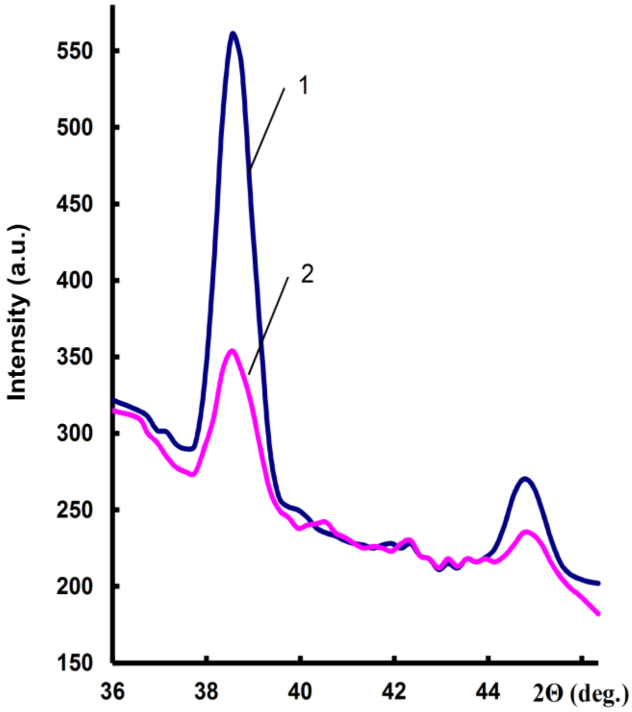

Fig.5 XRD diagram: 1 − without assistance; 2 − with assistance

The estimation values in our case are: w = 8.2x10^{15}, w_p = 5.7x10^{16}, Q = 2x10^{14} c-1, i.e. $w_p > w > Q$. Hence, the absorption factor is $A(w) \sim (w_p / w)^2$. It means that with such a relation between the incident wave frequency and the metal (aluminium) plasma parameters the absorption decreases.

Thus, an increase in the reflectance is related to a decrease in the losses connected with the absorption of electromagnetic radiation.

4. Conclusions

In this paper, we have investigated the consequences of using an assist Ar ion beam in the magnetron sputtering deposition of the Al films. The photometric studies have shown that the films deposited with ion assist are characterized by a 4-5% higher reflection factor in the 200-240 nm range compared to the films deposited without ion assist. The conducted TEM and AFM analyses of the films have revealed that the crystallite size and roughness change insignificantly. It means that the geometrical factor cannot be considered the reason for the enhanced reflectance. The X-ray diffraction (XRD) studies have shown that in the films produced by the ion-assisted deposition the value of microstress is 5 times higher than in the films deposited without ion assist. The microstress value is proportional to the concentration of the crystallites' point defects. In other words, the application of ion assist leads to an increase in the point defect concentration. Thus, an increase in the concentration of point defects in the deposited films results in their enhanced reflectance. It is connected with the change in the conditions of the electromagnetic wave interaction with matter.

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