

# The Surface Roughness Variation Due to Ion Irradiation During Thin Film Growth

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Experimentally observed non-monotonous dependence of thin films surface roughness on ion energy is analyzed by kinetic rate equation model. The modeling results show good quantitative agreement with the experiment and give possible physical reasons of behavior of the surface roughness. It is shown that non-monotonous dependence of surface roughness on ion energy is determined by the size of islands and diffusivity of atoms on the surface.

**Keywords:** Volmer-Weber growth, surface diffusion, kinetic modeling, surface roughness.

## 1. INTRODUCTION

In physical vapor deposition (PVD) the properties of thin films largely depend on the microstructure. The microstructure can be controlled by process parameters such as substrate temperature, working pressure, deposition rate, incident ion energy and flux, angle of incident particles, etc. Low energy ion beam systems form an essential part in various surface processing techniques such as ion beam etching, deposition and ion beam assisted growth. Muller [1] showed that energetic ion beam deposition increases adatom mobility at low temperature, resulting in denser microstructure and often to larger grain size. Higher incident ion energy provides an opportunity for surface diffusion reconstruction to obtain a smooth surface and a smaller average grain size [2]. R. Brahma and M.G. Krishna showed that the grain shape, size and their distribution on a substrate can be simply achieved by controlling the ion energy for a given flux [3]. The grains are flatter and exhibit elongated oblate shape at lower ion energy. As the incident ion energy is increased the rate of vapor arrival increases resulting in more spherical grains due to the higher impact rate and consequently shorter relaxation time.

The surface morphology of thin film is very much affected by ion irradiation during growth due to various ion-solid interactions. D. Kim [4] obtained that in a low energy region rms roughness of growing thin film increases proportionally with ion beam energy due to the growth of grain size by enhanced adatom mobility. However, in a high energy region the surface roughness decreases significantly. Since surface defects produced by intense ion beam bombardment act as nucleation centers and it favors the decrease of grain size. The non-monotonous dependence of surface roughness on incident ion energy was obtained and by other authors [5, 6] as well.

The purpose of this work is to give explanation of non-monotonous dependence of surface roughness on ion energy. The modeling results are obtained by a kinetic model, which is based on the rate equations and includes processes of surface diffusion, nucleation, growth and

coalescence of islands, sputtering in the case of thin film growth in Volmer-Weber mode. The theoretical results show a good quantitative agreement with experimental ones and allow explain the physical reason of non-monotonous surface roughness variation with ion energy.

## 2. KINETIC MODEL

The influence of ion energy on the surface roughness of thin films was analyzed by a kinetic equation model, which describes all steps of thin film growth. The model developed previously (detail description of the model can be found in our previous works [7]) has been improved taking into account the process of sputtering (etching). In the model it is assumed that the surface of substrate is atomically flat and the thin film grows in Volmer-Weber mode. The surface of growing thin film is bombarded by low energy ions. The sputtering yield  $Y_d$  is introduced to describe the sputtering process. Because of low energy of incident ions sputtering yield depends on ion energy linearly. In model, it is assumed that incident energetic species can remove the atoms from islands edge only. The islands dissociation due to the intense ion bombardment is not taken into account in this model.

All steps of deposition process are expressed in the system of the kinetic equations. These equations define the time variations of surface coverage  $\varphi$  and the islands density  $n$  during the thin film deposition. The coverage of the first monolayer of growing thin film is separated into coverage by single adatoms  $\varphi_S$  and coverage by clusters (islands)  $\varphi_C$ :

$$\begin{cases} \frac{d\varphi_S}{dt} = \alpha_{A0}i_0(1 - \varphi_L) - \alpha_{AA}i_0\varphi_S; \\ \frac{d\varphi_C}{dt} = 2 \cdot \alpha_{AA}i_0\varphi_S + a(t)\alpha_{AC}i_0\varphi_C^* + \\ \quad + B^{(1)}\alpha_{AC}i_0(1 - \varphi^{(1)})\left(\varphi^{(1)} - \varphi^{(2)}\right) - Y_d i_0\varphi_C; \\ \frac{d\varphi^{(K)}}{dt} = A^{(K)}\alpha_{AT}i_0\left(\varphi^{(K-1)} - \varphi^{(K)}\right) + \\ \quad + B^{(K)}\alpha_{AT}i_0\left(\varphi^{(K)} - \varphi^{(K+1)}\right)\left(\varphi^{(K-1)} - \varphi^{(K)}\right); \\ \frac{dn}{dt} = 0.5 \cdot \alpha_{AA} \cdot i_0\varphi_S + \alpha_{col}C_{col}(1 - \varphi_C - \varphi_\beta) \end{cases} \quad (1)$$

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The first equation presents the kinetics of the coverage of the first monolayer by the single atoms  $\varphi_S$ .  $i_0$  is the relative flux of arriving single atoms and  $(1 - \varphi_L)$  represents uncovered part of substrate which also involves diffusion length of single atoms  $\lambda$ . The coverage of the first monolayer increases when the arriving atom sticks to the substrate surface with the probability  $\alpha_{A0}$ .  $\varphi_S$  decreases due to the cluster nucleation process (the arriving atom sticks to the already existing single atom with the probability  $\alpha_{AA}$  and forms a new cluster of two atoms).

The second equation is used to describe the kinetics of the coverage of the first monolayer by the islands (clusters)  $\varphi_C$ . It increases after nucleation process (the first term) and when the single atom sticks at the edge of the island with the probability  $\alpha_{AC}$  (the second term). The probability  $\alpha_{AC}$  involves the Ehrlich-Schwoebel edge barrier.  $\varphi_C$  increases when the atoms jump down from the top of the island because of the surface diffusion process and sticks at the edge of the island (the third term).

The surface coverage by islands decreases when incident energetic species remove atoms from islands edge (the fourth term).  $\varphi_C^*$  is dimensionless area with radius  $\lambda$  around the clusters within which arrived atom will stick at the edge of cluster.  $a(t)$  is normalization factor [7]. Because of small ion energy sputtering yield  $Y_d$  has a linear dependence on ion energy.

The kinetics of  $K^{th} > 1$  monolayers  $\varphi^{(K)}$  coverage is described by the third equation. The coverage of  $K^{th}$  monolayer increases when atoms directly arrive on this monolayer and remain on it (the first term) or when the atoms jump down from the upper monolayer because of the surface diffusion (the second term). The possibility of the atom to stick on the given monolayer (i.e. on the surface of cluster) or to jump down from it is defined by the coefficients  $A$  and  $B$ , which depend on the coverage of monolayer and diffusivity of adatoms on the surface of island, defined by diffusion length  $\lambda_T$  (exact definition in ref. [7]).

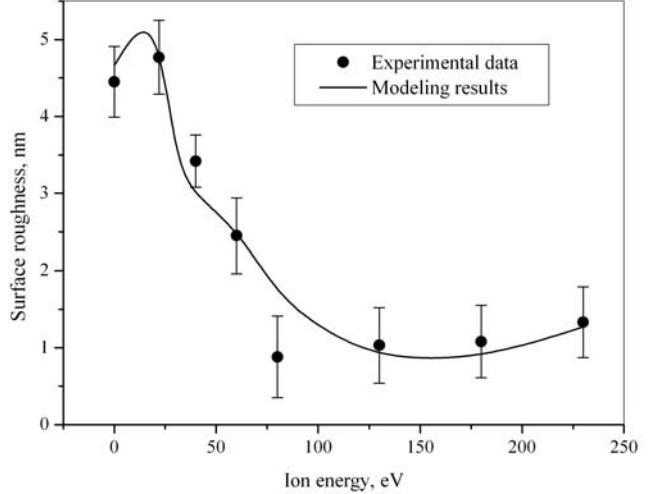
The fourth equation presents the kinetics of the relative islands density  $n$ . The island density increases and after nucleation (the first term). The island density decreases because of the coalescence effect (the second term). The islands are in a contact as they radius reaches the critical radius. It was assumed that the coalescence process stops at this moment. From that moment the number of the clusters remains constant. The process of the nucleation stops as well.

The surface roughness  $\delta(t)$  as a function of deposition time is defined as  $\delta(t) = K_2(t) - K_1(t)$  (in monolayers), where  $K_1(t)$  and  $K_2(t)$  are the number of monolayers with coverage  $\varphi^{(K_1)} = 0.06$  and  $\varphi^{(K_2)} = 0.94$ , respectively.

### 3. RESULTS AND DISCUSSIONS

The quantitative study of surface roughness variation as dependence on ion energy was performed using the proposed kinetic model in order to understand the influence of the ion energy. The modeling results were compared with the experimental ones obtained by Patsalas and coworkers [6] for TiN thin films deposited on  $\text{SiO}_2$  substrates. Thin films  $\sim 100$  nm were grown by

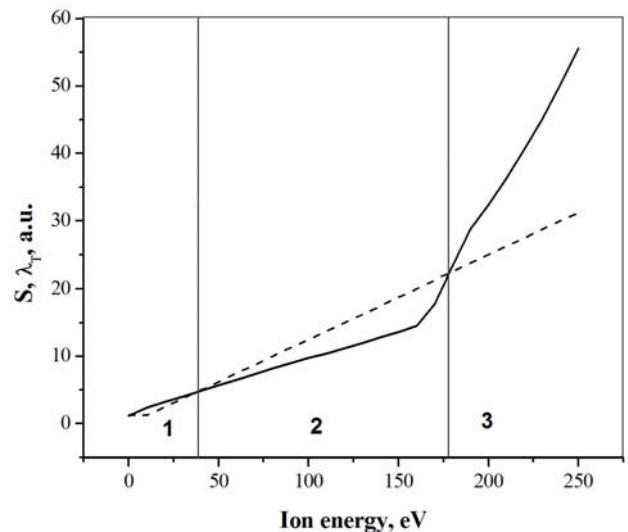
dc-unbalanced magnetron sputtering at room temperature. The experimental data show that the surface roughness dependence on ion energy is non-monotonous: surface roughness increases when incident ion energy is lower than 20 eV. In energy range of 20 eV–80 eV the surface roughness of thin TiN films decreases and when ion energy exceeds 80 eV it begins to increase again (Fig. 1, points).



**Fig. 1.** The dependence of the surface roughness on ion energy: experimental data [6] and modeling results. The coefficients used in calculations:  $\alpha_{A0} = 0.36$ ;  $\alpha_{AA} = 0.00035$ ;  $\alpha_{AC} = 0.095$ ;  $\alpha_{AT} = 0.95$ ;  $\alpha_{col} = 0.000065$ ;  $C = 1$ ;  $\beta = 18$ ;  $\lambda_T = \lambda - 1$ ;  $i_0 = 0.017$

The raising of ion energy increases the kinetic energy of adatoms. In model the kinetic energy (mobility) of adatoms interrelates with diffusion length. The adatom diffusion length on the substrate  $\lambda$ , diffusion radius on the top of islands  $\lambda_T$  and sputtering yield  $Y_d$  were varied during calculations. All other parameters were kept as constants.

The non-monotonous surface roughness dependence on ion energy could be explained by the interplay between the island size and the diffusion length of adatoms [8].



**Fig. 2.** Island size and diffusion length of adatom versus ion energy

At low ion energy the atoms, which drop on the top of island, remain on it, because, despite small size of island

the diffusion length of adatom is smaller and adatom cannot jump to lower layers (Fig. 2, region 1). The rougher surface is formed in this case.

With the increase of ion energy the diffusivity of adatom and size of island increase. However, the island size may increase not so fast as the diffusivity of adatom. The diffusion length exceeds sometimes the island size (Fig. 2, region 2). Atoms, which drop on the top of island, can jump down and fill the lower layer. The surface becomes smoother in this case. At higher ion energy the size of islands may exceeds the diffusion length again (Fig. 2, region 3). Adatoms remain on the top of islands and the surface roughness again increases.

The etching of surface island edges is negligible because of low ion energy. The process of subplantation which can be observed in a low range of incident ion energy has bigger influence on roughness of interface as on surface roughness of growing thin film. (similar results are obtained by authors of experiment [6]). Therefore the variation of surface roughness with increasing ion energy is exclusively attributed to the adatom mobility during growth.

## CONCLUSIONS

1. The non-monotonous dependence of surface roughness on incident ion energy can be explained in some degree by interplay between the islands size of growing thin film and adatoms diffusion length on the top of islands. If diffusion length exceeds the size of island, the surface roughness decreases and vice versa, when diffusion length is smaller as island size the surface roughness increases.
2. The sputtering of surface island edges is negligible because of low ion energy. Therefore non-monotonous

variation of surface roughness with increase of ion energy is exclusively attributed to the adatom mobility.

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