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Scanning Surface Roughness of Growing Nanoisland Thin Films as a Dependence on Substrate Temperature and Deposition Flux

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Experimentally observed non-monotonous dependencies of surface roughness of the growing thin films on substrate temperature and deposition flux of particles are analyzed by kinetic model based on rate equations. Obtained modeling results show good quantitative agreement with the experimental curves and explain so unusual phenomenon of surface roughness. It is shown that non-monotonous dependence of surface roughness on the temperature and deposition flux is determined by the size of islands and diffusivity of atoms on the surface. From the presented analysis follows that the formation mechanisms of non-monotonous dependencies of the surface roughness on the temperature and the deposition flux are different. These mechanisms are qualitatively analyzed in the present paper.
Keywords: rate equations, kinetic modeling, surface roughness, nanoclusters, island films, surface diffusion.

1. INTRODUCTION

The properties of the thin films are mostly determined by their microstructure. The microstructure of thin film depends on mechanisms of nanoclusters growth at initial stages, which can be analyzed by study of kinetics of surface roughness of growing film. Complicated dependencies of surface roughness on different technological parameters such as substrate temperature, ion energy, flux of arriving atoms, incident energy, deposition rate, etc. have been investigated experimentally by the many authors [1 – 11]. Yua et al [1] showed that the substrate temperature and energies of incident particles can promote the mobility of surface atoms and lead to smooth growth of films. M. Levlin and A. Laakso [2] studied the importance of different deposition rates at temperatures and found that higher temperatures roughen the films. Quantitative AFM analysis of flat titanium thin films showed a linear relation trend between the surface roughness and deposition flux [12]. There are many works where the non monotonous surface roughness dependencies on the substrate temperature [5 – 9] and deposition flux [13 – 16] are observed. J. Dumont et al [13] explain non-monotonous dependence of the surface roughness on the deposition rate by relation between the average distance between islands and critical island radius, at which a second layer nucleates on the top of these islands (the so called "TDT approach" theory). The layer-by-layer growth is observed (surface become smoother) if critical island radius is larger than the average distance between the islands. If critical island radius is small compared to the average distance between islands, the islands will nucleate a second layer before coalescence, giving multilayer growth (surface become rougher). By a three-dimensional kinetic Monte Carlo simulation technique [11] it was shown that with increase of the substrate temperature more adatoms can be activated to diffuse and fill in the inner voids of the film decreasing the surface roughness with temperature. However, when the temperature becomes too high, the diffusivity of adatoms is very high so that some of them can jump onto the top of layer. Then the surface roughness increases with temperature again.

The purpose of this work is to give explanation of non-monotonous dependencies of the surface roughness on substrate temperature and on the deposition flux. The curves are simulated by a kinetic model, which is based on the rate equations and includes processes of surface diffusion of the adatoms and the clusters, nucleation, growth and coalescence of islands in the case of thin film growth in Volmer-Weber mode. The theoretical results show a good quantitative agreement with the experimental results and allow to explain the physical reasons of the non-monotonous surface roughness variations with the substrate temperature and deposition flux.

2. KINETIC MODEL

The main idea of the model is to separate coverage of the first monolayer into coverage by single atoms φ_S and by the islands φ_C. It gives possibility to consider processes of islands nucleation and growth, coalescence and other kinetic processes. The set of model equations is following:

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

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All steps of deposition process are expressed by kinetic equations defining the time variation of surface coverage φ and cluster density n. The first, second and third equations describe the kinetics of surface coverage by single atoms φ_S, clusters of mean size φ_C and coverage of higher monolayers $\varphi^{(K)}$, $K>1$, respectively. The fourth equation describes the kinetics of island density n. i_0 is the relative flux of deposited atoms. $\alpha_{A0}\cdot\alpha_{AC}$ and α_{AC} are the sticking coefficients of adatoms to the substrate, to the edge of cluster and to the top of cluster, respectively. $\varphi_L=\varphi_S+\varphi_C+a(t)\varphi_C^*$ is the covered surface, where $\varphi_C^*=n\lambda^2+2\lambda\sqrt{n\varphi_C}$ is the dimensionless area with radius λ (diffusion length) around the cluster where adatoms can diffuse and stick at the edge of the cluster. α_{col} is the sticking coefficient of two clusters during coalescence process. $\varphi_\beta=n\beta^2+2\beta\sqrt{n\varphi_C}$ is the dimensionless area around the island with radius β were clusters can migrate (this term describes mobility coalescence process). The possibility of the atom to stick on the top of the cluster or to jump down from it is defined by the coefficients A and B, which depend on the coverage and diffusivity of adatoms on the top of island, defined by diffusion length λ_T. The detailed description of the model can be found in our previous work ref. [17, 18].

Diffusion lengths λ of adatom on the substrate and on the deposited material λ_T both depend on temperature. The diffusion length can be expressed as $\lambda=\sqrt{4Dt}$, where D is the diffusion coefficient, t is time. The diffusion length as a function of the substrate temperature $\lambda(T_{sub})$ can be expressed from Arrhenius law for diffusion as:

$$\lambda=2\sqrt{D_0\exp\left(\frac{-E_a}{kT_{sub}}\right)\cdot t}\,, \qquad (2)$$

which gives the increase of diffusion length with the substrate temperature, where D_0 is the preexponential term, E_a is the surface diffusion activation energy, k is Boltzmann's constant, T_{sub} is the substrate temperature.

Surface roughness $\delta(t)$ (in monolayers) as a function of deposition time t is defined as: $\delta(t)=K_2(t)-K_1(t)$, where $K_1(t)$ and $K_2(t)$ are numbers of the monolayer with the coverage equal to $\varphi(K_1(t))=0.05$ and $\varphi(K_2(t))=0.95$ respectively.

3. RESULTS AND DISCUSSIONS

3.1. Influence of substrate temperature

Many experimental results show that the dependence of surface roughness on the substrate temperature is non monotonous: the curves pass minimum and maximum points (see Figs 1 and 2 [7, 8]). This phenomenon was analyzed by the proposed model. According to the model, single atoms arrived to the surface of substrate diffuse until they find adsorption sites.

The diffusion length λ depends on temperature T_{sub} (eq. (2)). It was assumed that different substrate temperature influences the values of the diffusion lengths λ and λ_T only, other parameters were kept constant and independent on substrate temperature. The results obtained by modeling were quantitatively compared with the experimental ones obtained by S. G. Yoon et al. [7] for Ta_2O_5 thin films deposited on Si (111) substrates (Fig. 1). In Fig. 2 there are presented the quantitative comparison of modeling and experimental results obtained by P. Sobotik et al. [8] for Au thin films deposited on mica substrates. In both cases a good agreement between the experimental and modeling results is obtained.

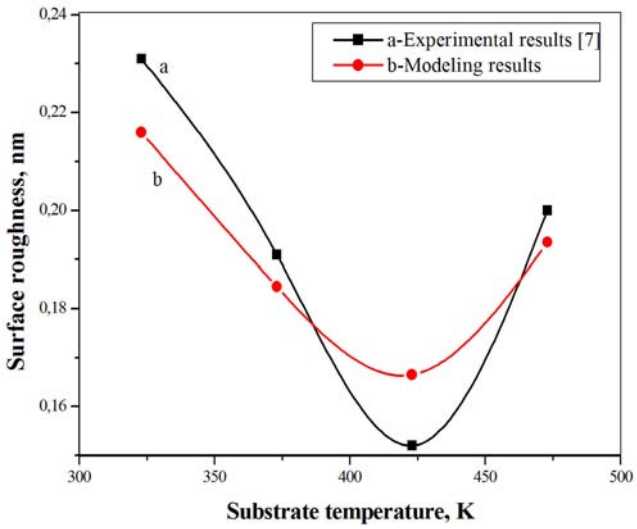

Fig. 1. Dependence of the surface roughness on substrate temperature. Curve: a – experimental results, Ta_2O_5 thin films deposition on Si (111) [7]; b – modeling results. The coefficients used in calculations: $\alpha_{A0}=0.1$; $\alpha_{AA}=0.0005$; $\alpha_{AC}=0.1$; $\alpha_{AT}=1.0$; $\alpha_{col}=0.0001$; $C=1$; $\beta=20$; $\lambda=\lambda_T$; $i_0=0.2$

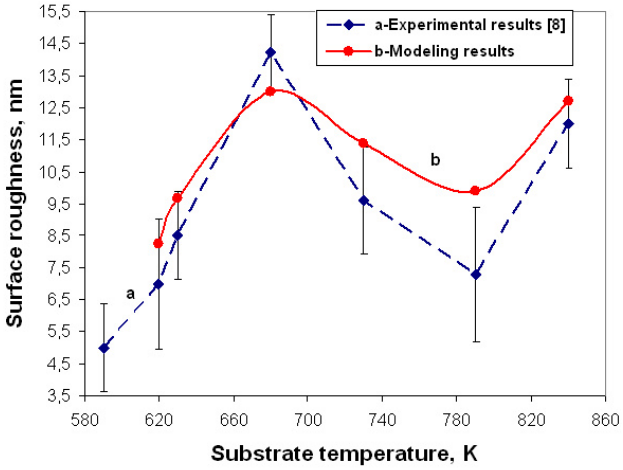

Fig. 2. The comparison of the dependencies of surface roughness on the substrate temperature obtained by a) experimental. [8] for Au thin films deposited on mica substrates and b) modeling results. $\alpha_{A0}=0.1$; $\alpha_{AA}=0.1$; $\alpha_{AC}=0.1$; $\alpha_{AT}=1.0$; $\alpha_{col}=0.0001$; $C=1$; $\beta=25$; $\lambda=\lambda_T$; $i_0=0.05$

The modeling shows that non-monotonous dependencies of the surface roughness on the substrate temperature occur because of interplay between the island size and the diffusion length of adatoms.

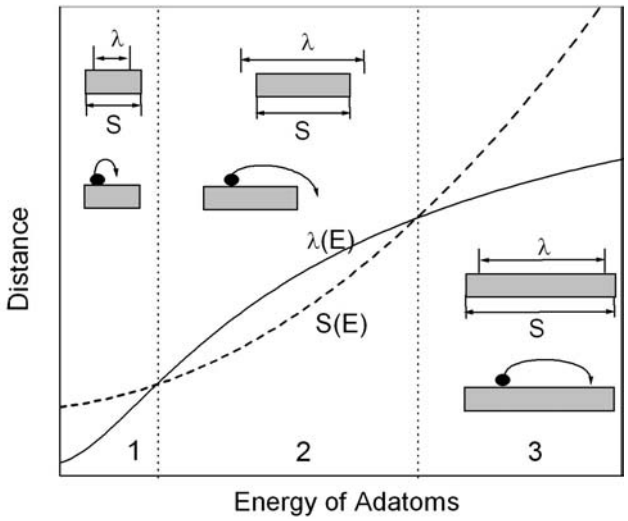

Fig. 3. Schematic draw of the functions of island size and diffusion length of adatoms and interplay between them forming non-monotonous dependence of surface roughness versus energy of adatoms

The schematic drawing of this phenomenon is shown in Fig. 3. At low temperatures both the diffusion length and island size are small.

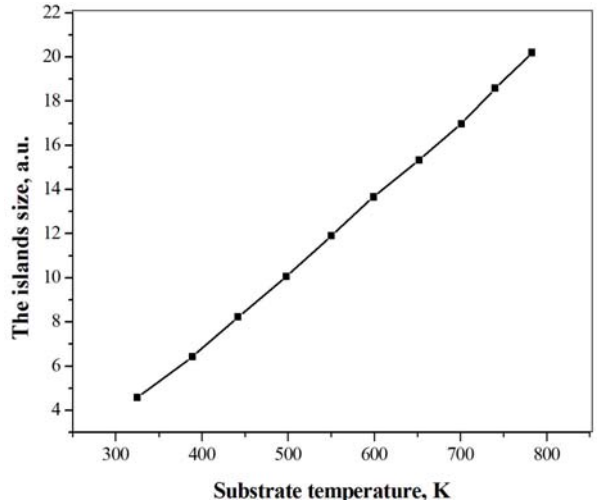

Fig. 4. The calculated dependence of island size on substrate temperature

The calculated dependence of island size on temperature is presented in Fig. 4, which shows continuous increase of island size with temperature. Despite the small island size, the diffusion length λ_T is too low at low temperatures and the adatoms arrived to the top of cluster stick on it (Fig. 3, regime 1). The formed surface in that case becomes rougher. The diffusivity of adatoms and the size of islands both increase with temperature but not linearly and with different speed (this is shown schematically in Fig. 3, regime 2).

It follows that the interval of temperatures may exist where diffusion length of adatoms exceeds the radius of islands. In that case the atoms arrived on the top of the islands jump down to the deeper layers. As a result the surface becomes smoother. With increase of temperature the island size again becomes higher than diffusion length and atomic jumps from the tops of island stop (Fig. 3, regime 3). The surface roughness starts to increase again.

3.2. Influence of flux

There are many experimental observations that surface roughness depends non-monotonously on flux of arriving particles. The flux of arriving particles does not affect diffusivity of atoms (considering number of particles not speed), and the explanation of this phenomenon must be different from the case of dependencies on temperature done above.

The study of the surface roughness of the growing thin film as the dependence on the flux of arriving atoms was performed using the proposed kinetic model in two cases:

1) diffusivities of atoms on surface of substrate λ and on the deposited material λ_T are the same ($\lambda = \lambda_T$);

2) diffusivity of atoms on the surface of substrate λ is significantly higher than the diffusivity of adatoms on deposited material λ_T($\lambda >> \lambda_T$).

It is well known that at random deposition (random sticking of atoms, no islands) the surface roughness increases with deposition time t as the function $\delta \sim \sqrt{t}$ [19]. The exact expression for the surface roughness can be found in that case as $\delta = \sqrt{4\alpha\, i_0 t}$ [20], where α is the sticking coefficient and i_0 is the relative atomic flux. From this expression it is seen that the flux influences surface roughness similarly as deposition time: surface roughness δ increases with the flux of arriving atoms i_0 as the function of $\delta \sim \sqrt{i_0}$.

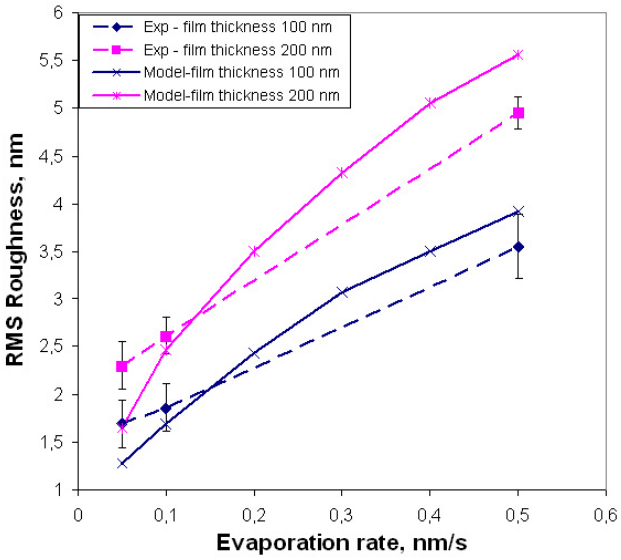

Fig. 5. The dependence of the surface roughness on the flux of arriving atoms in the case $\lambda = \lambda_T$. The dashed lines are experimental results of Ti flat thin film growth on glass substrates [12] and solid lines are the modeling results. The coefficients used in calculations: $\alpha_{A0} = 0.05$; $\alpha_{AA} = 0.005$; $\alpha_{AC} = 0.1$; $\alpha_{AT} = 1.0$; $\alpha_{col} = 0.001$; $C = 1$; $\beta = 15$; $\lambda = \lambda_T = 2$

The analysis of flux influence on the surface roughness was performed assuming that the flux of arriving atoms does not affect the sticking and diffusivity of adatoms. Only value of atomic flux in model equations

was changed in these calculations. The calculated dependencies of the surface roughness on the deposition flux in the case of $\lambda = \lambda_T$ are shown in Fig. 5. The results are compared with the experimental data [12] for Ti flat thin film growth. The calculated curves follow the law of $\delta \sim \sqrt{i_0}$, i. e. the surface roughness monotonously increases with the deposition flux in this case ($\lambda = \lambda_T$). However, the law $\delta \sim \sqrt{i_0}$ is valid for the case of random sticking (no islands) of arriving atoms only.

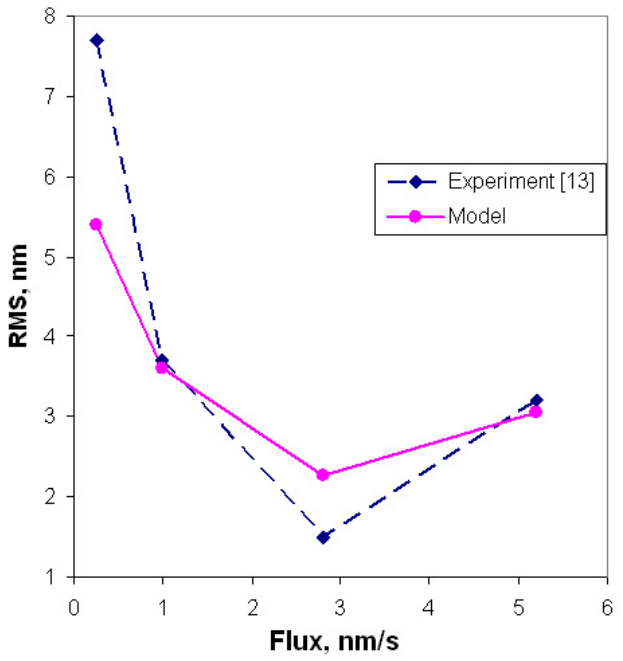

Fig. 6. The dependence of surface roughness on the flux of arriving atoms in the case $\lambda >> \lambda_T$. Experimental for thin silver films deposited on mica substrate [13] and modeling results. The coefficients used in calculations: $\alpha_{A0} = 0.05$; $\alpha_{AA} = 0.001$; $\alpha_{AC} = 0.1$; $\alpha_{AT} = 1.0$; $\alpha_{col} = 0.0001$; $C = 1$; $\beta = 25$; $\lambda = 30$; $\lambda_T = 1$

The deposition with island formation can be considered as driving deposition, which means that adsorption of atoms is influenced by some driving forces under which geometrical structures (islands) are formed.

Completely different dependence of surface roughness on the deposition flux is observed in the study of J. Dumont et al. [13] for thin silver films deposited on mica substrate shown in Fig. 6. The experimental dependence is not monotonous: decreasing at the low deposition fluxes and increasing at the higher fluxes. Taking into model values of diffusion length which fulfill the condition $\lambda >> \lambda_T$ the same behavior is obtained by model calculations. The calculated results are presented in Fig. 6 and show a good quantitative agreement.

As follows from the presented model the decrease of the roughness with increase of the deposition flux occurs because of atomic jumps from the top of islands when diffusion length exceeds the size of island (regime 1 in Fig. 3). As it is shown in Fig. 7, where calculated dependencies of the island size on the deposition flux are presented, the size of islands decreases with increase of the deposition flux. Despite both curves $\lambda = \lambda_T$ and $\lambda >> \lambda_T$ show the same behavior, the island size in the case $\lambda >> \lambda_T$ is significant higher and with increase of flux, decreases faster.

In the case of larger islands, the decrease of the island size gives more intensive jumps of atoms from the islands to the substrate. As a result, the surface roughness decreases. However, the random sticking (described by the law $\delta \sim \sqrt{i_0}$) prevails again when islands become small.

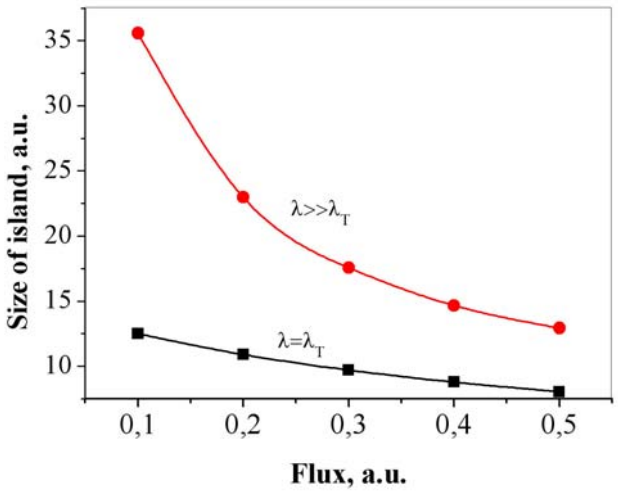

Fig. 7. The dependencies of the island size on the deposition flux in two cases $\lambda = \lambda_T$ and $\lambda >> \lambda_T$

The surface roughness then starts to increase as it is seen in Fig. 6 and in this part show the same behavior as results shown in Fig. 5. Random sticking always takes place which tends to increase the roughness and driving sticking is just additional term which in the case of large islands reduces this tendency.

4. CONCLUSIONS

1. The explanation of non-monotonous dependence of surface roughness on the temperature is following: i) at low temperatures the surface roughness decreases with temperature because of small island size and relatively large diffusion length. The atomic jumps of arriving atoms from the top of islands prevails; ii) at higher temperatures island size becomes larger and exceeds the diffusion length. The arriving atoms stick on the top of islands and the surface roughness increases with the temperature.
2. The explanation of non-monotonous dependence of surface roughness on the deposition flux is following: there is general rule that surface roughness at the random deposition (no islands) increases with deposition flux as function of $\delta \sim \sqrt{i_0}$. However, there is deviation from this rule if the relatively large islands (comparing with diffusion length of adatoms) are formed: the arrived atoms jump down from the top of island and the decrease of the surface roughness is observed. However, island size decreases with increasing of deposition flux and at higher fluxes, very small islands are formed. In that regime the jumps of atoms from the top of islands are not important any more and the random deposition regime prevails at which surface roughness starts to increase with deposition flux.

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