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REGULAR ARTICLE

Influence of Interactions between Adsorbate and Substrate on the Statistical Properties of Surface Structures at Condensation

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We investigate the effects of elastic adsorbate-substrate interactions on the kinetics of nano-structuring of thin films during condensation and statistical properties of the surface structures in the framework of theoretical approaches and numerical simulations. We derive the reaction-diffusion model for the spatiotemporal evolution of the adsorbate concentration on the substrate by taking into account adsorption, de-sorption, transferring of adatoms between neighbor layers, adsorbate-substrate and adsorbate-adsorbate interactions and lateral diffusion. By exploiting stability analysis of the stationary homogeneous states to in-homogeneous perturbations we have shown that an increase in the elastic adsorbate-substrate interaction strength induces formation of stable adsorbate structures on the substrate under low-pressure condensation. We perform numerical simulations of the processes of adsorbate structures formation during deposition with varying adsorbate-substrate interaction strengths. It is found that an increase in the strength of elastic ad-sorbate-substrate interactions accelerates processes of adsorbate self-organization into stable clusters and leads to the formation of larger number of adsorbate islands with elevated sizes. It is shown that in the stationary regime the distribution of stable adsorbate islands over sizes remains universal for different values of the elastic interaction strength between adsorbate and substrate. This study provides valuable insights into the mechanisms governing nano-structured thin film growth in low-pressure systems with varying elastic adsorbate-substrate bonding strengths by depositing different materials onto different substrates.

Keywords: Adsorptive systems, Nano-structured thin films, Surface structures, Adsorbate-substrate interaction, Numerical simulations

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1. INTRODUCTION

Innovative nano-structured thin films play a crucial role in cutting-edge advancements in transistors [1], energy harvesting [2], sensors [3], and catalysts [4]. Among various fabrication techniques, nano-structured thin films grown via low-pressure deposition methods have attracted considerable attention due to their versatile applications in electronics, optics, catalysis, and sensing technologies [5]. Precise control over their key properties, such as morphology, crystallinity, and surface chemistry, is essential for optimizing their performance. A fundamental aspect of achieving this control lies in understanding the influence of adsorbate-substrate interactions on film growth dynamics [6].

Adsorbate-substrate interactions encompass a broad range of physical and chemical phenomena that govern the initial nucleation, growth kinetics, and final structural characteristics of thin films. These interactions are dictated by factors such as surface energetics, lattice matching, van der Waals forces, and chemical bonding configurations [7]. Strong interactions promote ordered nucleation and the formation of well-defined islands or clusters. The nature and intensity of these interactions are critical in determining the film growth mode – whether it follows a layer-by-layer (Frank-van der Merwe) or island (Volmer-Weber) mechanism [8].

Mathematical and numerical modeling of nano-structured thin-film growth provides valuable insights into the underlying dynamics. These models allow for the systematic investigation of key factors, including chamber pressure, deposition temperature, energy characteristics, and external influences, all of which affect the morphology, type and size of surface structures during growth. A widely employed approach in this field involves reaction-diffusion models [9-12], which offer predictive insights that facilitate the fine-tuning of technological parameters to achieve the de-sired physical and chemical properties in thin films. Notably, competition between atomic deposition and reaction processes can lead to the stabilization of nanoscale spatial patterns, even in monoatomic layers [13]. These patterns may serve as templates for subsequent stages of film texture evolution. Representative examples of such

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systems include Al or Cu layers deposited on Si substrates, as well as SiO_2 and TiN layers on Ti or Al substrates [10].

In this study, we conduct a computational analysis of the evolution of a monoatomic layer deposited on a substrate under low-pressure condensation, employing a continuous dynamical reaction-diffusion model. We explore the impact of adsorbate-substrate interactions on the formation and statistical properties of adsorbate structures. Our findings demonstrate that increasing the strength of these interactions can induce adsorbate patterning in the first growing layer and govern statistical properties of surface structures.

The work is organized in the following way. In the next Section we construct the reaction-diffusion model of the spatiotemporal evolution of adsorbate on the first growing layer during low-pressure condensation from gaseous phase by taking into account adsorption, desorption, transference between layers, lateral diffusion adsorbate-substrate and adsorbate-adsorbate interactions. In Section 3 we perform stability analysis of the homogeneous states to inhomogeneous perturbations in order to define the range of control parameters (adsorption coefficient, adsorbate-substrate and adsorbate-adsorbate interaction strengths) when the separate stable adsorbate structures can be formed on the substrate during condensation. In Section 4 we present results of numerical simulations of the nano-structuring of thin films during deposition and discuss an influence of adsorbate-substrate interaction strength onto dynamics of pattern formation and statistical properties of surface structures. We conclude in the last Section.

2. MATHEMATICAL MODEL

We consider a two-layer model describing the spatiotemporal evolution of adsorbate on a substrate during condensation in adsorptive systems. Our analysis assumes the deposition of a single atomic species and utilizes a reaction-diffusion model to track the local adsorbate concentration, $x_1(\mathbf{r}, t) \in [0, 1]$, within a unit cell of size 1 on the first growing layer. Following the approach outlined in Refs. [14], we define the concentration on the second layer as:

$$x_2(x_1) = \left((x_1)^{1/2} - \beta\right)^2 \tag{1}$$

where β represents the dimensionless terrace width of multi-layer structures.

In general, the reaction-diffusion equation governing the field $x_1(\mathbf{r}, t)$ takes the form:

$$\partial_t x(\mathbf{r}, t) = R(x_1, x_2) - \nabla \cdot \mathbf{J}(x_1) + \xi(\mathbf{r}, t) \tag{2}$$

where the reaction term $R(x_1, x_2)$ accounts for adsorption, desorption, and transference of adatoms between adjacent layers. The adsorbate flux $\mathbf{J}$ is defined by the free energy $\mathbf{F}$ of the adsorbed layer:

$$\mathbf{J}(x_1) = - D_0 M(x_1) \nabla \delta\mathbf{F}/\delta x_1. \tag{3}$$

Here, D_0 is the diffusion coefficient, and $M(x_1) = x_1 (1 - x_1)$ ensures that diffusion occurs only on available sites; $\nabla = \partial/\partial_r$. The last term in Eq. (2), $\xi(\mathbf{r}, t)$, represents a stochastic source accounting for microscopic effects related to adatom redistribution, which is modeled as white zero-mean delta-correlated Gaussian noise with the small intensity σ^2:

$$\langle \xi(\mathbf{r}, t) \rangle = 0;$$
$$\langle \xi(\mathbf{r}, t)\xi(\mathbf{r}', t') \rangle = \sigma^2 \, \delta(t - t')(r - r').$$

The reaction term governing the first adsorptive layer is expressed as $R(x_1) = R_a(x_1) + R_d(x_1) + R_t(x_1)$. Adsorption is described by: $R_a = k_a(1 - x_1)(1 - x_2)$, where the adsorption rate is given by $k_a = \varpi p \exp(- E_a / k_B T)$, with E_a being the adsorption energy, ϖ the frequency factor, p the gas-phase pressure, k_B the Boltzmann constant, and T the temperature. Adsorption requires available sites on both the first and second layers. Desorption is governed by $R_d = - \varpi \, x_1(1 - x_2) \exp(U(x_1) / k_B T)$, where $U(x_1)$ is the interaction potential accounting for adsorbate-adsorbate and adsorbate-substrate interactions. Interlayer transfer reactions are captured by $R_t = k_t(x_2 - x_1)$, which describes the reduction in adsorbate concentration due to transfer between layers.

The free energy $\mathbf{F}$ for the first adsorptive layer, as given in Refs. [9-12], is:

$$\mathbf{F} = \int \mathrm{d}r \, [f_0(x_1) + (k_B T)^{-1} x_1 U(x_1)]. \tag{4}$$

Here, the entropic non-interactive contribution is: $f_0(x_1) = x_1 \, ln(x_1) + (1 - x_1) \, ln(1 - x_1)$. The interaction potential $U(x_1)$ includes contributions from adsorbate-substrate interactions, $U_{el} = - \varepsilon_s$ (where $\varepsilon_s > 0$) represents attractive interactions), and adsorbate-adsorbate interactions, $U_{ads}(x_1)$. The adsorbate interaction potential is defined by: $U_{ads}(r) = - \int \mathrm{d}r' \, u(r - r')x(r')$. Here, the binary attractive potential $u(r)$ is assumed to be of symmetric form such that $\int r^{2m+1} u(r) \, \mathrm{d}r = 0$, for $m = 1, 2, \ldots$. Following Refs. [9-12], we approximate $u(r)$ using a Gaussian profile:

$$u(r) = 2\varepsilon_b (4\pi r_0{}^2)^{-1} exp(- r^2/(4r_0{}^2)), \tag{5}$$

where ε_b is the interaction strength and r_0 is the interaction radius. Assuming minimal variation of adsorbate concentration within the interaction radius r_0, we apply a self-consistent approximation widely used in numerical modeling [10-17]:

$$\int \mathrm{d}r' u(r - r')x(r') \approx \int \mathrm{d}r' u(r - r') \, \Sigma_m [(r - r')^m / m!] \nabla^m x(r).$$

By substituting Eq. (5) and assuming $r_0{}^{2m} \to 0$ for $m \geq 2$, the adsorbate interaction potential simplifies to: $U_{ads}(r) = - 2\varepsilon_b(x_1 + r_0{}^2 \nabla^2 x_1)$.

The term U_{el} becomes particularly significant when adatom-substrate bonding is strong, as in the case of Cu atoms deposited on Ti, Ta, or Mo substrates. However, for systems with minimal lattice mismatch, such as Al deposition on TiN (where mismatch is ~ 4 %), elastic stress effects can be neglected. From Eq. (3), it follows that these interactions do not influence the lateral flux, $\mathbf{J} \propto \nabla U$. However, in systems with large lattice mismatches, strong bonding suppresses desorption during the initial growth phase, as desorption rate depends on the interaction potential $U(x_1)$.

To facilitate analysis, we introduce dimensionless parameters $\alpha = k_a / \varpi$ and $\gamma = k_t / \varpi$, scaling time by the adatom lifetime $\tau_d = \varpi^{-1}$. The spatial coordinate r is normalized by l, while the diffusion length is given by $L_d^2 = D_0 / \varpi$, with $L_d = 20r_0$ and $l = 4r_0$. Combining all reaction and diffusion terms, Eq. (2) for $x = x_1$ reads:

$$\begin{aligned} \partial_t x(\mathbf{r}, t) = \alpha(1 - x)(1 - x_2(x)) - \\ - x(1 - x_2(x))\, exp(- \varepsilon(2x + \delta)) + \gamma(x_2(x) - x) + \\ + D_l^2[\nabla^2 x - 2\varepsilon\nabla M(x)\cdot\nabla(x + \rho^2\nabla^2 x)] + \xi(\mathbf{r}, t). \end{aligned} \quad (6)$$

Here $\varepsilon = \varepsilon_b / k_B T$, $D_l = L_d / l$ and $\rho = r_0 / l$. This study focuses on the effects of interaction strength ε_s on the kinetics of thin-film structuring and the morphology of the first growing layer. To this end, we introduce $\delta = \varepsilon_s / \varepsilon_b$ as the primary control parameter, with $\gamma = 0.1$, $\beta = 0.05$ and $\sigma^2 = 0.01$ fixed throughout our analysis.

3. STABILITY ANALYSIS

To determine the role of adsorbate-substrate interactions, characterized by the strength δ, in the growth of nano-structured thin films and their impact on surface morphology, we first establish the conditions necessary for the formation of stable adsorbate structures on the substrate during deposition. To this end, we employ a standard stability analysis of stationary homogeneous states x_{st} against inhomogeneous perturbations. The homogeneous stationary state can be found from the Eq. (6) by taking $\partial_t x = 0$ and $\nabla x = 0$. We consider a small deviation of the adsorbate concentration $x(\mathbf{r})$ from its homogeneous state x_{st}, represented as $\delta_x(\mathbf{r}) = x(\mathbf{r}) - x_{st} \propto e^{\lambda t + ik\mathbf{r}}$, where k is the wave number, and $\lambda(k)$ is the stability exponent. This assumption leads to $\partial_t\delta_x = \lambda(k)\delta_x$ and $\nabla\delta_x = ik\,\delta_x$. Applying linear stability analysis, we expand the reaction term around x_{st}, neglecting higher-order terms δ_x^n for $n > 1$. From Eq. (6), we obtain the expression for the stability exponent:

$$\lambda(k) = d_x R(x)\,|_{\,x = xst} - D_l k^2[1 - 2\varepsilon M(x_{st})(1 - \rho^2 k^2)]. \quad (7)$$

If $\lambda(k) < 0$ for all k, then the system remains in a stable homogeneous state x_{st}. This implies that during deposition, the adsorbate will cover the substrate uniformly, preventing the formation of distinct surface patterns. Conversely, if $\lambda(k) > 0$ for certain wave numbers $k \in (k_1, k_2)$, spatial instabilities will grow over time, leading to the emergence of stable surface patterns on the substrate. By analyzing $\lambda(k)$ for different values of the control parameters, we can determine the parameter ranges where pattern formation occurs.

Previous studies have shown that under high-pressure condensation, an increase in the adsorption rate leads to a re-entrant scenario of pattern formation and changes in surface morphology [12]. In contrast, our focus is on the ability of elastic interactions between the adsorbate and substrate to induce pattern formation during low-pressure condensation. The corresponding stability diagram $\delta(\varepsilon)$, illustrating the domain of pattern formation, is presented in Fig. 1: domain I corresponds to the homogeneous coverage of the substrate by adsorbate; domain II relates to the system parameters, when separate stable adsorbate structures will be formed on the substrate during deposition.

The results indicate that at low adsorption rates (corresponding to low-pressure condensation), increasing the adsorbate-substrate interaction strength δ induces the formation of stable surface structures at a critical value $\delta = \delta_c$. An increase in the adsorbate-adsorbate interaction strength ε leads to a decrease in the critical value δ_c. A comparison of the curves in Fig. 1 reveals that reducing the adsorption coefficient α necessitates higher values of both the elastic interaction strength δ and the interaction strength ε to achieve stable pattern formation during condensation.

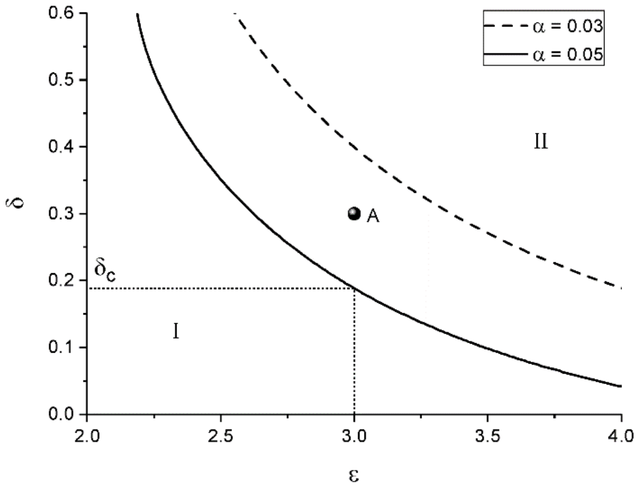

Fig. 1 – Stability diagram of stationary homogeneous states subjected to inhomogeneous perturbations at various values of the adsorption coefficient α

Next, we fix $\varepsilon = 3$ and $\alpha = 0.05$ and vary the elastic interaction strength δ in order to characterize an influence of the adsorbate-substrate interaction strength δ onto statistical properties of stable adsorbate structures formed on the substrate in the framework of numerical simulations.

4. RESULTS OF NUMERICAL SIMULATIONS

To investigate the dynamics of adsorbate island formation on the first growing layer, we perform numerical simulations by solving Eq. (6) on a two-dimensional hexagonal grid. The system has a linear size of $L = N\Delta x$, where $N = 512$ represents the number of sites, and the spatial integration step is set to $\Delta x = 0.5$. The temporal evolution is computed using a time step of $\Delta t = 10^{-3}$ till the dimensionless time $t = 10^3$. Spatial derivatives are evaluated using the Fourier spectral method [18], ensuring accurate numerical differentiation. The initial condition assumes a clean substrate, meaning the adsorbate concentration is set to $x(\mathbf{r}, 0) = 0$. To maintain continuity in the simulation domain, periodic boundary conditions are applied. Snapshots of the typical evolution of the surface morphology during condensation at $\delta = 0.3$ (point A in Fig. 1) are shown in Fig. 2. The adsorbate concentration is represented using a grey-scale, where black corresponds to $x(\mathbf{r}) = 0$ (no adsorbate) and white corresponds to $x(\mathbf{r}) = 1$ (full coverage). During the condensation process, the adsorbate islands begin to organize after an incubation period, t_c. As deposition continues, the number of islands increases, and their size grows accordingly.

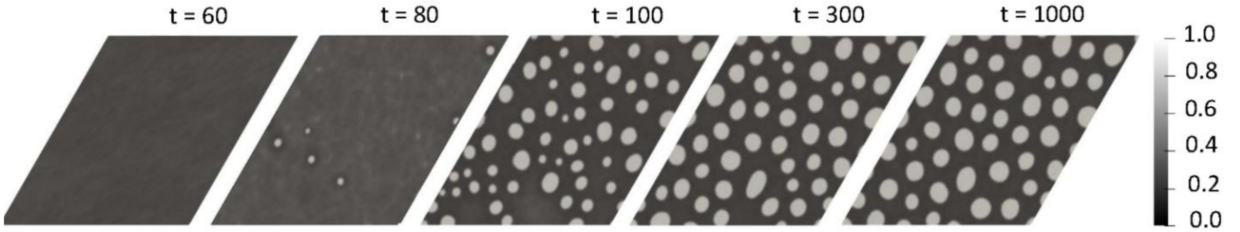

Fig. 2 – Snapshots of the typical evolution of the surface morphology during condensation at $\delta = 0.3$ (point A in Fig. 1) at different time instants. Here snapshots are shown for the linear size of the computational grid $N = 256$

In Fig. 3, we present the evolution of the mean radius of adsorbate structures $\langle R \rangle$, scaled by the length l for different values of the adsorbate-substrate interaction strength δ. The linear size of an adsorbate structure is associated with the radius of a circle having the same area. The time dependencies mean number density $\langle N \rangle$, scaled in units of area of simulation grid N^2 of absorbate structures is shown in the inset in Fig. 3. The number of adsorbate structures and the area of each structure were calculated by exploiting the machine learning method DBSCAN (Density-based spatial clustering of applications with noise) [19] by using a threshold $\phi_{th} = 0.5$ in order to distinguish the adsorbate phase from the matrix (substrate) phase. This method allows one to count the number of adsorbate clusters and their population (area). In order to perform averaging five independent experiments for the same set of control parameters were done. It is observed that after the incubation time t_c, both quantities increase rapidly during the growth stage. The mean number density $\langle N \rangle$ of adsorbate structures reaches a maximum value before starting to decrease, while the growth rate of $\langle R \rangle$ slows down, entering the coarsening stage. After long-term condensation, the adsorbate morphology reaches a quasi-stationary state, characterized by N_{st} adsorbate structures with a mean linear size R_{st}. From results shown in Fig. 3 one sees, that an increase in the interaction strength δ between substrate and adsorbate accelerates processes of adsorbate structures formation by decreasing the incubation time t_c. Moreover, with increase in δ both the number density of adsorbate structures and their mean size increase.

Next, let us discuss an influence of the adsorbate-substrate interaction strength onto dynamics of adsorbate structures formation and their statistical properties. To that end we wary δ in the interval [0.2, 0.5] and analyze the incubation time t_c and stationary values of the mean size of adsorbate structures R_{st} and their number density N_{st} after long-term condensation. The dependencies $t_c(\delta)$,$R_{st}(\delta)$ and $N_{st}(\delta)$ are shown in Fig. 4. It is seen, that $\delta > \delta_c$ (see Fig. 1) the incubation time t_c decreases rapidly (see circles in Fig. 4) and the number density N_{st} (see triangles in Fig. 4) increases fast with the interaction strength δ growth.

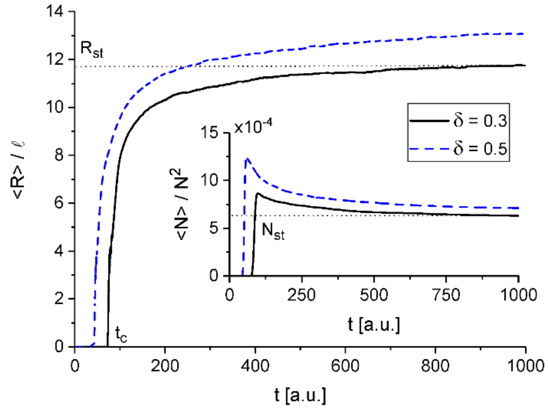

Fig. 3 – Evolution of the mean radius of adsorbate structures ⟨R⟩ (and their mean number density $\langle N \rangle$ shown in inset) at $\delta = 0.3$ and $\delta = 0.5$

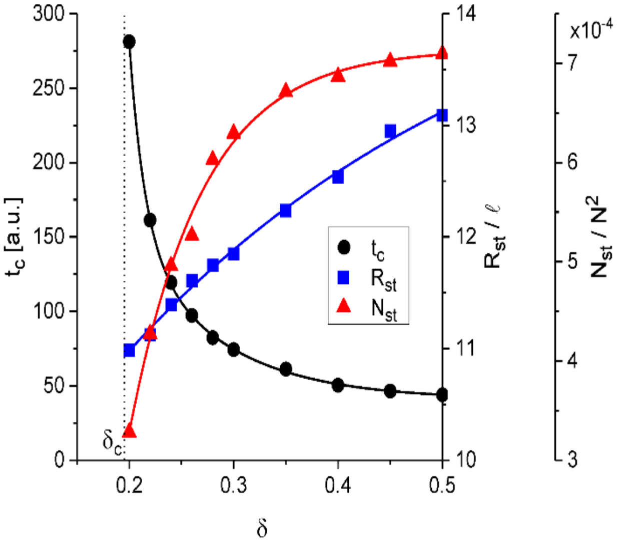

Fig. 4 – Dependencies of the incubation time t_c and stationary values of the mean size of adsorbate structures R_{st} and their number density N_{st} after long-term condensation on the adsorbate-substrate interaction strength δ

At elevated values of δ both these quantities tend to saturation. The stationary value of the mean linear size of adsorbate structures R_{st} monotonically grows with increase in the interaction strength δ (see squares in Fig. 4). Hence, by choosing different substrate and/or adsorbed spices (different interaction strength between adsorbate and substrate) one can control number and size of adsorbate structures on the substrate.

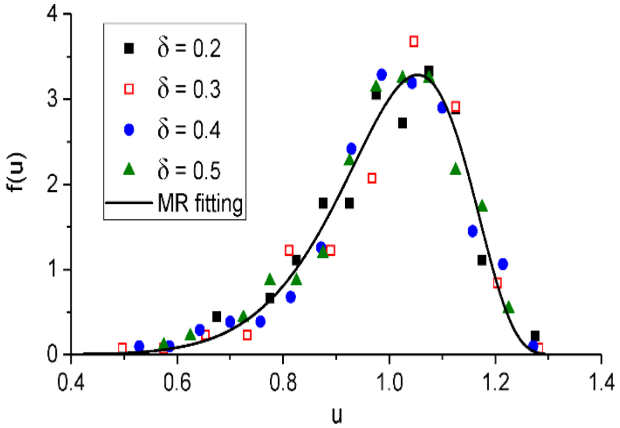

Fig. 5 – The distribution of adsorbate structures over sizes $f(u)$, where $u = R / \langle R \rangle$, at different values of the adsorbate-substrate interaction strength δ at final stage ($t = 10^3$)

Finally, let us analyze the distribution of adsorbate structures over sizes $f(u)$, where $u = R / \langle R \rangle$, at different values of the adsorbate-substrate interaction strength δ at final stage of condensation ($t = 10^3$), shown in Fig. 5. It follows that the obtain normalized numerical distribution (shown by different markers) is of universal character independent on the interaction strength δ and corresponds well to the Marquesee and Ross (MR) approach [20] shown by solid curve.

5. CONCLUSION

In this study, we have investigated the effects of adsorbate-substrate interactions on the morphology of growing thin films during low-pressure condensation using numerical simulations. Our results demonstrate that at such conditions, these interactions induce the self-organization of adatoms, leading to the formation of separate adsorbate islands on the substrate. We found that stronger adsorbate-substrate interactions accelerate the formation of adsorbate structures, resulting in an elevated number of larger adsorbate clusters.

In summary, adsorbate-substrate interactions play a crucial and multifaceted role in nano-structured thin film growth under low-pressure deposition conditions. A deeper understanding and precise control of these interactions are essential for tailoring thin film properties to meet specific application requirements.

The general findings of this work provide valuable insights for predicting surface morphology during low-pressure condensation and optimizing technological conditions to fabricate nano-structured thin films with desired morphologies and statistical properties. The dimensionless parameters used in this study, including adsorption rate and adsorbate-adsorbate and adsorbate-substrate interaction energies, can be rescaled for specific adsorbate-substrate systems using experimental data.

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Вплив взаємодії між адсорбованими частинками та підкладкою на статистичні властивості поверхневих структур при конденсації

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У роботі досліджено вплив пружної взаємодії між адсорбатом і субстратом на кінетику наноструктурування тонких плівок під час конденсації та статистичні властивості поверхневих структур в рамках теоретичних підходів і чисельних симуляцій. Розроблено модель реакційно-дифузійного типу для просторово-часової еволюції концентрації адсорбату на підкладці, яка враховує адсорбцію, десорбцію, переходи адатомів між сусідніми шарами, взаємодію адсорбат-субстрат і адсорбат-адсорбат, а також поверхневу дифузію. Використовуючи аналіз стійкості стаціонарних однорідних станів до неоднорід-них збурень показано, що збільшення сили пружної взаємодії між адсорбатом і підкладкою індукує формування стійких структур адсорбату на підкладці при конденсації в умовах низького тиску. Дина-міку процесів формування структур адсорбату під час осадження з різними значеннями сили взаємодії адсорбат-субстрат досліджено в рамках процедури числового моделювання. Виявлено, що збільшення сили пружної взаємодії між адсорбатом і субстратом прискорює процеси самоорганізації адсорбату з формуванням стійких кластерів та приводить до формування більшої кількості островів адсорбату з більшими розмірами. Показано, що в стаціонарному режимі розподіл стійких островів адсорбату за розмірами залишається універсальним для різних значень сили пружної взаємодії між адсорбатом і субстратом. Проведені дослідження надають цінні відомості про механізми, які керують ростом наноструктурованих тонких плівок у при конденсації в умовах низького тиску при різних значеннях сили пружної взаємодії між адсорбатом і підкладкою при осадженні різних матеріалів на різні субстрати.

Ключові слова: Адсорбційні системи, Наноструктуровані тонкі плівки, Поверхневі структури, Взаємодія адсорбат–підкладка, Чисельне моделювання.