THE FORMATION KINETICS OF THIN-FILM COATINGS UNDER HEAVY-ION BOMBARDMENT

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A model describing the initial stage of the nucleation and evolution of thin-film coatings under the bombardment of the surface with an ion beam has been proposed. The evolution equations for the surface profile and the distribution function of absorption centers have been obtained, as well as the quasistationary solutions of these equations in the case of a weakly non-uniform distribution of absorption centers and a flat profile of the surface at the initial moment. The conditions for the nuclei to be created in the form of a spherical dome have been determined in terms of the model parameters.

1. Introduction

A large number of applied problems in surface science is related to the study of processes of formation and growth of deposited films on a solid surface. The film substance can be either specially sputtered or sorbed from the gas environment onto the substrate surface. In many experiments, an electric field, which promotes the deposition of ionized particles, and the irradiation of the surface with an ion beam (the ion-beam-assisted deposition, IBAD) are in use.

The profile of the surface changes in the course of material sputtering onto it. In particular, the evolution of the film made up of the deposited material passes through the stage of formation of nuclei or "islands" (the island stage). It is evident that, for such islands to be formed, adsorption (accommodation) centers for particles of a deposited material have to exist on the surface at the initial moment. These accommodation centers are distributed over the surface in accordance with some law which can change during the surface evolution. In particular, as was mentioned above, the deposition may be accompanied by the ion-beam bombardment of the surface. In addition to the sputtering of the surface (including the particles deposited on it), the ion beam can stimulate the formation of new accommodation centers. The latter circumstance should be beneficial for the film growing on the surface (which has, in the general case, a rather complicated profile). In its turn, an increase of the thickness of a deposited film should "screen" the accommodation centers that are available on the surface, thus slowing down the processes of film nucleation. Therefore, we deal with several processes which are connected to each other in a complex nonlinear manner.

The results of experimental researches of the formation of film textures at initial stages show unequivocally that the process concerned is ion-stimulated. Irrespective of how ions stimulate the deposition processes, we can call two mechanisms that are responsible for a change of the character of grain formation under the action of irradiation [1]. These are (i) the formation of defects on the substrate surface and, at later stages of the film growth, on the surface of the film itself and (ii) the direct penetration of metal atoms with a low energy into the subsurface layers of the substrate. The formations of both kinds become additional centers of accommodation. The first and second mechanisms are more characteristic, respectively, of the implantation-assisted deposition and the magnetron sputtering.

All this results in the nucleation of grains, which is more intense in comparison with the case of thermal deposition [2], on the one hand, and the activation of the process stimulated by the irradiation-induced surface diffusion, on the other hand. As a consequence, the grained structure, which is being formed under the ion irradiation, becomes more equiaxial, and its columnar structure disappears even at low temperatures of the substrate [3]. In addition, the double distribution of nuclei over their dimension [1] is typical of this stage of the nucleation under irradiation. It is a consequence of the processes that run simultaneously: namely, the surface diffusion of metal atoms, coalescence of small grains, and nucleation of new ones on released sites.

Other effects of the surface activation upon the ion irradiation are a reduction of the epitaxy temperature of a deposited metal and the formation of a structure with densely packed planes parallel to the substrate [4–6].

Among a plenty of theoretical works devoted to the subject under consideration (see, e.g., works [7, 8]), many works describe the formation of the surface relief while depositing various materials onto it upon both the presence and the absence of sputtering processes (see, e.g., works [9–15]).

In the present work, we propose a promising, in our opinion, approach to the description of the evolution of the surface profile that is formed by particles of the deposited material if the surface is irradiated with an ion beam. The model is based upon rather general assumptions concerning the processes that take place and the parameters that describe the surface. The model allows a simple derivation of the evolution equations for the mentioned parameters. For the verification of the model, the stationary (quasistationary) solutions of the evolution equations, obtained for the case of an isotropic, weakly nonhomogeneous distribution of the accommodation centers at the initial moment, have been analyzed. It has been shown that, under certain conditions, the stationary (quasistationary) profile of the surface looks like a plane which includes islands possessing the form of a spherical dome (in this connection, see work [15]).

2. Basic Points of the Model

The basic postulate of the suggested model is an assumption that the evolution of the surface is described by two characteristics (the description parameters) at any moment. One of those characteristics is the distribution function of adsorption centers (accommodation centers) over the surface. The other is the surface profile. Under conditions when the relief is formed by deposited particles of one kind, the surface profile characterizes the thickness of the film above a certain point of the substrate.

The surface profile at the time t will be described by the function $z(x, y, t)$ which establishes the dependence of the coordinate z (the applicate) on two other Cartesian coordinates x and y (the abscissa and the ordinate, respectively) on the $x0y$ plane, where 0 is the origin of the coordinate system. The positive direction of the z axis is reckoned upwards from the $x0y$ plane.

We suppose that the particles consisting of only one material are deposited onto the surface of the substrate from an atomizer or the residual atmosphere. In this work, we are interested in the very process of the growth of new thin-film formations which is connected with the adsorption of particles of the deposited material. Therefore, for the sake of definiteness, we assume hereafter that

$$z\left(x, y, t\right) - z_0\left(x, y\right) \geq 0, \qquad z_0\left(x, y\right) \geq 0, \tag{1}$$

where $z_0\left(x, y\right) = z\left(x, y, t = 0\right)$ is the profile of the substrate surface at the initial moment. If the process of growth of new formations at the expense of deposited particles occurs under the irradiation of the surface with an ion beam, condition (1) evidently reflects the prevalence of the deposition over the sputtering at every point of the surface.

Let us define now the distribution function $f(x, y, z(x, y))$ of accommodation centers over the solid surface which has the profile $z(x, y, t)$. In this work, the distribution function $F(x, y, z, t)$ means the density of accommodation centers at a point with coordinates (x, y, z) within the elementary volume $dV = dxdydz$ at the moment t. If the objects are arranged on an arbitrary surface with profile $z(x, y)$, the distribution function $F(x, y, z)$ would obviously look like

$$F\left(x, y, z\right) = f\left(x, y, z\right) \delta\left(z - z\left(x, y\right)\right)$$

or

$$F\left(x, y, z\right) = f\left(x, y, z\left(x, y\right)\right) \delta\left(z - z\left(x, y\right)\right), \tag{2}$$

where $\delta\left(z\right)$ is the Dirac delta-function. The function $f\left(x, y, z\left(x, y\right)\right)$ defined by formula (2) is the desired distribution function of adsorption (accommodation) centers on the surface of a solid. Its physical sense consists in that the quantity $f(x, y, z(x, y))dxdy$ equals a number of accommodation centers around a point $z(x, y)$, the projections of which onto the $x0y$ plane lie within an elementary area $dS = dxdy$. Since the integral of the function $F(x, y.z)$ over the whole volume has to be equal to the total number of accommodation centers N,

$$\int\limits_V dVF\left(x, y, z\right) = N,$$

the normalization of the distribution function $f(x, y, z(x, y))$ is determined in accordance with Eq. (2) by the formula

$$\int dx \int dy f\left(x, y, z\left(x, y\right)\right) = N, \tag{3}$$

where the integration is carried out over the whole $x0y$ plane.

The quantities introduced into consideration – the surface profile $z(x, y, t)$ and the distribution function of accommodation centers $f(x, y, z(x, y, t))$ – are the basic characteristics which describe the surface modification in the framework of the proposed model. While deriving the evolution equations for these characteristics, one must take into the account the possibility for the deposited substance to be sputtered with the help of the ion beam and then redeposited at other points of the surface. The effects induced by the crystal structure of the substrate will not be taken into account in this work.

3. The Equation of the Surface Evolution

The rate of the surface profile modification $\dot{z}(x, y, t)$, according to the basics of the model, is governed by two terms:

$$\dot{z}(x, y, t) = S_1(x, y, t) + S_2(x, y, t)\,. \tag{4}$$

These terms, $S_1(x, y, t)$ and $S_2(x, y, t)$, describe the contributions of the competing processes of deposition of the particles of the deposited material onto the surface and sputtering them from it, respectively, by the ion beam.

Let us specify firstly the form of the quantity $S_2(x, y, t)$ which defines the rate of the surface modification due to its sputtering as follows [11–14]:

$$S_2(x, y, t) = YvJ_2\cos\vartheta(x, y, t)\,. \tag{5}$$

Here, J_2 is the flux density of primary ions in the beam that bombards the surface (in this work, it is considered uniform; see below), Y the sputtering factor, and v the characteristic volume of the substance which corresponds to one removed particle (one should bear in mind that the matter concerns the substance that is deposited onto the surface). By its order of magnitude, this volume can be estimated as that of a cube, the linear dimensions of which are about the size of the atom of the deposited material.

The angle $\vartheta(x, y, t)$ in expression (5) is the angle between the z axis and the normal to the surface at a point (x, y):

$$\cos\vartheta(x, y, t) = \left\{1 + \left(\frac{\partial z}{\partial x}\right)^2 + \left(\frac{\partial z}{\partial y}\right)^2\right\}^{-1/2}. \tag{6}$$

Some explanations for expression (5) are needed, because the quantity $S_2(x, y, t)$ has a rather complex form in the general case. It is known that the sputtering factor Y is defined as the number of detached particles per one ion of the beam impinging on the surface. For this reason, it is an integrated quantity which has a clear physical sense only in the case of the sputtering of a plane surface, all points of which are equivalent. There are numerous experiments devoted to the measurement of the sputtering factors of various materials (in this connection, see work [11]). Just in the case of plane surfaces, these experiments allow one to establish certain regularities that characterize the dependence of the sputtering factor on the beam energy, the relationship between the masses of the target particle and the particle which sputters the surface, and so on. It is evident that, when considering the sputtering of a surface with developed relief, it is necessary to introduce, instead of the integral characteristic Y, some specific quantity. The last characterizes the surface sputtering at a point of observation $[x, y, z(x, y)]$, provided that a sputtering ion strikes the surface at a point $[x_0, y_0, z(x_0, y_0)]$ and at an angle $\vartheta_1(x_0, y_0)$ to the normal of the profile $z(x_0, y_0)$. However, the definition of such a quantity is a separate, rather complicated problem. Until now, the solution of this problem is sought independently in every specific case in the framework of model ideas which are convenient for this case only but cannot be united with other ideas in the framework of a more general approach. For this reason, for the sputtering factor Y in expression (5), we adopt the value that is averaged over the angle of incidence and over the range of characteristic energies of the primary beam for the plane surface.

Another important characteristic of the sputtering process, which governs the magnitude and the direction of the surface erosion rate, is the distribution of sputtered particles over the coordinates. To a great extent, the same characteristic also defines the process of redeposition of the particles that were knocked out of the surface. The majority of works, where the spatial distributions of sputtered particles were studied, was devoted to the measurement of the dependence of the sputtered particle yield on the polar angle reckoned from a normal to the surface of a plane polycrystalline target [11]. However, it is clear that the sputtering of a surface with arbitrary relief would produce the yield of secondary particles, the angular distribution of which would differ essentially from that obtained in the case of a plane target.

In what follows, we are interested in the sputtering processes induced by rather small flux densities J_2. In this case, the sputtering process should not suppress the process of formation of relief textures from the deposited material on the substrate surface. Analyzing the experimental data concerning the sputtering of plane targets, one can draw a conclusion that upon the low flux densities of the impinging beams and the low energies of ions (from 1 to 10 keV), the maximum in the angular distribution of the yield of the secondary particles which are connected to the cascade processes of sputtering is reached at small polar angles. The position of this maximum weakly depends on the angle of incidence of the primary beam at such energies [11]. For this reason, one can consider, as the main approximation, that the maximal fraction of the secondary particle yield at every point of the surface is contained within a small solid angle, the polar axis of which is directed along the normal to the surface at this point. This circumstance will be used by us while accounting the processes of redeposition onto the developed relief of the surface.

Consider now the term $S_1(x,y,t)$ in Eq. (4), assuming it responsible for the processes of material deposition onto the substrate surface. We write down this term as

$$S_1(x,y,t) = \\ = v\alpha(x,y;z(x',y'),f(x',y'))\,J_1(x,y;z(x',y')), \qquad (7)$$

where $f(x,y;z(x',y',t))$ is the distribution function of accommodation centers over the profile surface $z(x,y,t)$ which is defined by formulae (2) and (3); $\alpha(x,y;z,f)$ is the probability of accommodation of a deposited particle at a point $[x,y,z(x,y)]$; and $J_1(x,y;z(x',y'))$ the flux density of particles that are deposited onto the surface at the point $[x,y,z(x,y)]$. The functional dependences of these quantities on the surface profile and the distribution function of accommodation centers reflect the circumstance that the state of the surface, in the framework of our model, must be described completely at any moment just by the functions $f(x,y;z(x',y',t))$ and $z(x,y,t)$. In particular, the functional dependence of the deposited particle flux $J_1(x,y;z(x',y'))$ on the surface profile may mean, e.g., that redeposited particles, i.e. the particles of the deposited material from other sections of the surface, can make an essential contribution to the deposition of particles onto the surface. As was mentioned above, the contribution of the particle redeposition can be substantial only provided the developed relief of the surface. For this reason, we may assume that, at $z(x,y,t) = \mathrm{const}(x,y)$,

$$J_1(x,y;z(x\prime,y\prime))\,|_{z=\mathrm{const}} = J_1(x,y),$$

where $J_1(x,y)$ is the flux density of the deposited substance from an evaporator or, may be, from the residual atmosphere. If these fluxes are isotropic, it is obvious that

$$J_1(x,y;z(x\prime,y\prime))\,|_{z=\mathrm{const}} = J_1 = \mathrm{const}(x,y). \qquad (8)$$

The latter expression will be used below, assuming that the flux of the deposited material is directed perpendicularly to the plane surface $z=0$.

Let us obtain now the evolution equation for the distribution function of accommodation centers $f(x,y;z(x,y))$. It is natural to assume that the variation rate of this distribution function in time, $\dot f(x,y;z(x,y))$, is determined by the state of the surface at the moment t. But the characteristics of the substrate surface, in the framework of the model proposed, are the distribution function itself and the state of the surface relief which is determined by the function $z(x,y,t)$ at the same moment t. Mathematically, those considerations can be expressed in terms of the functional dependence of the variation rate of the distribution function of accommodation centers $S_f(x,y,t)$ on the quantities $f(x,y;z(x,y))$ and $z(x,y,t)$:

$$S_f(x,y,t) = S_f(x,y;z(x',y'),f(x',y;z(x',y'))).$$

Thus, the evolution equation for the distribution function of accommodation centers can be written down as

$$\dot f(x,y;z(x,y)) = \\ = S_f(x,y;z(x',y'),f(x',y;z(x',y'))) \qquad (9)$$

in rather a general case.

Equations (3) and (9) together with expressions (4)–(7) constitute the system of equations which describe a modification of the surface when the deposition of atoms onto it from the gas phase or the bombarding by a beam of ions takes place. It is obvious that there is no practical use of this system until the explicit forms of the functionals $J_1(x,y;z(x',y'))$ and $S_f(x,y;z(x',y'),f(x',y;z(x',y')))$, as well as that of the accommodation probability α (see Eqs. (7) and (9)), will have been specified.

However, the problem of determination of these functionals, starting from the theoretical or experimental conceptions about the researched phenomena, has not been solved in the general form until now. For this reason, in order to study the initial stages of the growth of new formations, we expand the functionals $J_1(x,y;z)$ and $S_f(x,y;z,f)$ into functional series in the vicinity of the initial surface profile $z_0(x,y) = z(x,y,t=0)$ and the initial distribution function of accommodation centers $f_0(x,y;z)\,|_{t=0}$, and confine ourselves to the linear approximations:

$$J_1(x,y;z) \approx J_1(x,y;z_0) + \\ + \int_{-\infty}^{\infty} dx' \int_{-\infty}^{\infty} dy' I_z(x,y;x',y')\{z(x',y') - z_0(x',y')\},$$

$$S_f(x,y;z,f) \approx S_f(x,y;z_0,f_0) + \int_{-\infty}^{\infty} dx' \int_{-\infty}^{\infty} dy' K_z(x,y;x',y') \{z(x',y') - z_0(x',y')\} + \int_{-\infty}^{\infty} dx' \int_{-\infty}^{\infty} dy' K_f(x,y;x',y') \times \{f(x',y';z(x',y')) - f_0(x',y')\}, \quad (10)$$

where

$$I_z(x,y;x',y') = \left.\frac{\delta J_1(x,y;z)}{\delta z(x',y')}\right|_{z=z_0},$$

$$K_z(x,y;x',y') = \left.\frac{\delta S_f(x,y;z,f)}{\delta z(x',y')}\right|_{\substack{z=z_0\\ f=f_0}},$$

$$K_f(x,y;x',y') = \left.\frac{\delta S_f(x,y;z,f)}{\delta z(x',y')}\right|_{\substack{z=z_0\\ f=f_0}}. \quad (11)$$

The case where the surface profile at the initial moment is a plane $z_0(x,y) = \text{const}$ (if the origin of coordinates belongs to this plane, then $z_0(x,y) = 0$), should be discussed separately. In this case, according to Eq. (8), the quantity $J_1(x,y;z_0)$ in formula (10) does not depend on coordinates and coincides, in the main approximation, with the flux density J_1 of particles that are deposited onto the surface from either an evaporator or the atmosphere:

$$J_1(x,y;z=0) = J_1. \quad (12)$$

For the same reason, the quantity I_z in Eqs. (10) and (11) has to depend, in this case, only on the coordinate difference:

$$I_z(x,y;x',y') \equiv I_z(x-y';y-y'). \quad (13)$$

The formulae similar to Eq. (13) are also valid for the quantities K_z and K_f from expressions (10) and (11) at $z_0(x,y) = 0$:

$$K_z(x,y;x',y') = K_z(x-x';y-y'), \quad (14)$$

$$K_f(x,y;x',y') = K_f(x-x';y-y').$$

The reason why these expressions are valid lies in the fact that the variation rate of the distribution function of accommodation centers should not depend on coordinates in the case of the plane surface and the uniform distribution of accommodation centers over it.

It follows from formulae (10)—(14) that the rates of the processes under consideration at the point of observation (x,y) and at the moment t are governed, generally speaking, by the behavior of the surface state characteristics $z(x,y)$ and $f(x,y;z(x,y))$ at every other point of the surface at the same moment. However, it is natural to admit that the influence of these quantities on the rates of the processes running is maximal if the set of points (x',y') (see Eqs. (13) and (14)) are located near the point of observation (x,y). This means that the functions $I_z(x-x',y-y')$, $K_z(x-x',y-y')$, and $K_f(x-x',y-y')$ (see expressions (10)—(14)) have to possess sharp maxima at $x = x', y = y'$. Therefore, we may believe that the main contribution to the integrals over x' and y' in expressions (10) are given by the values of the parameters z, f, and f_0 at the point of observation (x,y). For this reason, expressions (10), in the main approximation and provided $z_0(x,y) = 0$, read

$$J_1(x,y;z) \approx J_1 + I_z z(x,y), \quad (15)$$

$$S_f(x,y;z,f) \approx S_f(x,y;f_0) - K_z z(x,y) - K_f\{f(x,y;z) - f_0(x,y)\}, \quad (16)$$

where the notations [see expressions (11), (13), and (14)]

$$I_z \equiv \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy I_z(x,y),$$

$$K_z \equiv \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy K_z(x,y),$$

$$K_f \equiv \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy K_f(x,y),$$

$$I_z > 0, \quad K_z > 0, \quad K_f > 0. \quad (17)$$

are introduced.

The signs of the quantities I_z, K_z, and K_f in formulae (15) and (17) were chosen from the following considerations. The second term on the right-hand side of expression (15) can be naturally interpreted as a contribution of the redeposition processes to the particle flux. From this point of view, the positiveness of the factor I_z is obvious. If the rate of generation of new accommodation centers is supposed to be higher on the surface that is free from deposited particles (for example, as a consequence of the bombarding action of the ion beam), the surface profile thickening caused by deposited particles should reduce the rate of generation of new accommodation centers. It is this circumstance that determines the positive value of the factor K_z and the choice of the minus sign before it in expression (16). It is natural to interpret the term $-K_f f(x,y;z)$ in formula (16) as a reduction of the number of accommodation centers per unit time owing to external factors, i.e. to those not connected with the development of the relief. For example, the development of cascade processes, which are stimulated by the bombardment of the surface with an ion beam, can be attributed to such factors. For this reason, the factor K_f must be positive. We also note that, in this case, the sum of terms $S_f(x,y;f_0)$ and $K_f f_0(x,y)$ in Eq. (16),

$$S(x,y) = S_f(x,y;f_0) + K_f f_0(x,y), \qquad (18)$$

has the meaning of the quantity that characterizes the rate of generation of new accommodation centers on the plane surface $z_0(x,y) = 0$, free from deposited particles.

The probability of the accommodation $\alpha(x,y;z)$ of particles deposited onto the substrate surface, similarly to the quantity S_f, is the functional of the surface characteristics in our model: the surface profile $z(x,y,t)$ and the distribution function of accommodation centers $f(x,y;z(x,y))$ (see Eq. (7)). Repeating the considerations and calculations that brought us to formulae (16)—(18) for the function S_f, we obtain

$$\alpha(x,y;z(x',y'),f(x',y')) \approx$$

$$\approx \alpha_0(x,y) - a_z z(x,y) + a_f f(x,y;z(x,y)), \qquad (19)$$

where

$$\alpha_0(x,y) = \alpha(x,y;0,f_0) - a_f f_0(x,y) \qquad (20)$$

and

$$a_z = -\int\limits_{-\infty}^{\infty} dx \int\limits_{-\infty}^{\infty} dy a_z(x,y) \geq 0,$$

$$a_f = -\int\limits_{-\infty}^{\infty} dx \int\limits_{-\infty}^{\infty} dy a_f(x,y) \geq 0. \qquad (21)$$

The functions $a_z(x,y)$ and $a_f(x,y)$ in formulae (21) look like

$$a_z(x,y) = \left.\frac{\delta\alpha(x,y;z,f)}{\delta z(x',y')}\right|_{\substack{z=0,\\ f=f_0}},$$

$$a_f(x,y) = \left.\frac{\delta\alpha(x,y;z,f)}{\delta f(x',y')}\right|_{\substack{z=0,\\ f=f_0}}. \qquad (22)$$

The signs of the factors a_z and a_f are chosen according to the following reasons. It is evident that the closer a deposited particle to the accommodation center, the higher should be the probability of accommodation. Therefore, the growth of the surface profile screens, as if, the accommodation center, by removing the as-deposited particles away from this center and, hence, by reducing the probability of accommodation. This circumstance is reflected by the minus sign before the second term on the right-hand side of expression (19) and by the sign of the factor a_z in Eq. (21). The positiveness of the factor a_f in Eqs. (19) and (20) is obvious, because, as the density of accommodation centers grows, the probability of accommodation of a particle deposited onto the surface has to increase. We note that the quantity $\alpha_0(x,y)$ (see Eq. (19)) is the probability of accommodation of a particle deposited onto the surface free from other deposited particles.

Thus, formulae (10)—(22) define, in the main approximation, the functionals that enter into the evolution equations (4) and (9). We emphasize that the quantities, which enter into Eqs. (10)—(22) (e.g., I_z, K_f, a_z, etc.) and which are easy to be given a certain physical sense, are the parameters of the theory and cannot be calculated in the framework of the proposed model.

Again, substituting expressions (5)—(7), (15), and (19) into Eqs. (4) and (8) and taking into account the terms linear in z and f bring us to the following system of equations which describes the evolution of the surface:

$$\dot{z}(x,y,t) = vJ_1\alpha_0(x,y) +$$

$$+v(I_z\alpha_0(x,y) - a_zJ_1)z(x,y,t) + va_fJ_1f(x,y;z) -$$

$$-vYJ_2\left\{1 + \left(\frac{\partial z}{\partial x}\right)^2 + \left(\frac{\partial z}{\partial y}\right)^2\right\}^{-1/2}, \qquad (23)$$

$$\dot{f}(x,y,t) = S(x,y) - K_z z(x,y,t) - K_f f(x,y,t). \qquad (24)$$

It should be noted that formulae (15)–(19) suppose slow variations of the functions $z(x,y)$, $f(x,y)$, and $f_0(x,y)$ over the coordinates at the dimensions of spatial inhomogeneities L which are characteristic of the processes under considerations:

$$L\left|\frac{\partial z}{\partial x}\right| \ll z, \quad L\left|\frac{\partial f}{\partial x}\right| \ll f, \quad L\left|\frac{\partial f_0}{\partial x}\right| \ll f_0 \ldots. \qquad (25)$$

For example, in the case where there are no distinguished directions of spatial inhomogeneities on the surface, expression (15) looks like

$$J_1(x,y;z) \approx J_1 + I_z z(x,y) + +D_z\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right) z(x,y) + ..., \quad (26)$$

where

$$D_z = \frac{1}{3}\int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy\left(x^2+y^2\right) I_z(x,y). \quad (27)$$

Taking a term such as the last indicated one on the right-hand side of Eq. (26) into consideration can be considered, in some sense, to be equivalent to making allowance for the processes of surface diffusion in the equations of the surface evolution. However, if the gradients of the parameters involved in the surface description are small (see Eq. (25)), the terms of this type may be neglected, as it was done in the present work. The account of such terms results in essential mathematical difficulties when solving the equations of the surface evolution analytically. In what follows, we are going to deal with the numerical solution of the evolution equations, such as Eqs. (22), taking into account the processes of surface diffusion.

System (22) is rather complicated and, generally speaking, cannot be solved analytically even in the case where the explicit expressions for the functions $\alpha_0(x,y)$ and $S(x,y)$ are known. The situation becomes somewhat simpler if we are interested only in the stationary (or quasistationary) solutions of Eqs. (22), i.e. in the asymptotical solutions at $t \to \infty$.

4. Stationary Solutions of the Surface Evolution Equations

For the sake of definiteness, we use the term "stationary" for the solutions throughout this section, although all calculations presented here are applicable to the determination of quasistationary solutions as well.

In the stationary case ($\dot{z} = 0$ and $\dot{f} = 0$), Eq. (24) is readily solved with respect to f:

$$f(x,y;z) = \left(S(x,y) - K_z z(x,y)\right)/K_f. \quad (28)$$

Substituting expression (28) into Eq. (23) at $\dot{z} = 0$, we obtain the equation for the determination of the stationary surface profile which was formed by the particles of the deposited material:

$$\Lambda(x,y) + \lambda(x,y) z(x,y) = = Y J_2\left\{1+\left(\frac{\partial z}{\partial x}\right)^2+\left(\frac{\partial z}{\partial y}\right)^2\right\}^{-1/2}. \quad (29)$$

Here, the following notations were introduced [see Eqs. (15)–(18), (20), and (21)]:

$$\Lambda(x,y) \equiv J_1\alpha_0(x,y) + \frac{a_f J_1}{K_f} S(x,y),$$

$$\lambda(x,y) \equiv I_z\alpha_0(x,y) - a_z J_1 - a_f J_1\frac{K_z}{K_f}. \quad (30)$$

For Eq. (29) to be solved in the general form, the explicit expressions for the functions $\Lambda(x,y)$ and $\lambda(x,y)$ are required. However, it can be shown that, even in the case where the initial distribution of accommodation centers is close to the spatially uniform one, Eq. (29) may have a solution that describes a developed relief formed by the particles of a deposited material (we recall that the surface was considered planar at the initial moment). Let the distribution function of accommodation centers at the initial moment, $f_0(x,y)$, have a single weakly pronounced maximum at a point (x_0,y_0) and the axial symmetry. Then, we may admit that

$$f_0(x,y) = f_0(x-x_0, y-y_0) \equiv f_0(\rho), \quad (31)$$

where

$$\rho \equiv \sqrt{(x-x_0)^2+(y-y_0)^2}. \quad (32)$$

In so doing, we actually pass to the polar coordinate system with the origin at the (x_0,y_0) point. It is natural to admit that, in this case, the functions $\Lambda(x,y)$ and $\lambda(x,y)$ [see Eqs. (15)—(18), (20), (21), and (30)] also depend on the coordinates (x,y) only in terms of ρ,

$$\Lambda(x,y) \equiv \Lambda(\rho), \quad \lambda(x,y) \equiv \lambda(\rho). \quad (33)$$

Hence, a solution of Eq. (29) should be sought in the form

$$z(x,y) \equiv z(\rho). \quad (34)$$

Furthermore, let us suppose that the distribution function $f_0(\rho)$ decays along with increase in ρ so slowly that conditions (25) are fulfilled for it. A slow decay of the distribution function as ρ grows, together with the existence of a weakly pronounced maximum at the point (x_0,y_0), does mean that the initial distribution of accommodation centers is close to a spatially uniform one. In accordance with that, we may suppose that, in the main approximation, the quantities $\Lambda(\rho)$ and $\lambda(\rho)$ (see Eq. (33)) are quite independent of ρ:

$$\Lambda(\rho) \equiv \Lambda = \text{const}, \quad \lambda(\rho) \equiv \lambda = \text{const}. \tag{35}$$

Since we are interested, first of all, in the formation of islands from the deposited material on the substrate surface, a solution of Eq. (29), taking into account formulae (31)—(35), is to be sought in the form of a solitary protrusion against the background of an equilibrium plane surface:

$$z(\rho) = z_1(\rho)\theta(R-\rho) + z_2\theta(\rho-R), \tag{36}$$

where

$$\theta(x) = \begin{cases} 0, & x<0 \\ 1, & x>0 \end{cases}, \tag{37}$$

is the unit function and R the radius of this protrusion's base which will be determined below. The function $z_1(\rho)$ in expression (36) describes the profile of a solitary protrusion against the background of the equilibrium plane surface at a level $z_2 = \text{const} \geq 0$, and the inequality [see expression (1)]

$$z_1(\rho)\theta(R-\rho) - z_2 \geq 0, \tag{38}$$

which is the condition for a developed relief to exist, has to be satisfied.

According to formulae (36) and (37), Eq. (29) is divided into the equation for the surface of a stationary protrusion,

$$\Lambda + \lambda z_1(\rho) = \frac{YJ_2}{\sqrt{1+\left(\frac{dz(\rho)}{d\rho}\right)^2}}, \quad \rho<R \tag{39}$$

and the equation for a stationary level of the plane surface $z_2 \geq 0$,

$$\Lambda + \lambda_1 z_2 = YJ_2, \quad \rho>R, \tag{40}$$

where (see Eqs. (17), (20), (30), (33), and (35))

$$\lambda_1 = \lambda - \alpha_0 I_z = -J_1\left(a_z + \frac{K_z}{K_f}a_f\right) < 0. \tag{41}$$

A new constant $\lambda_1 = \lambda(I_z=0)$ introduced in Eq. (40) reflects the absence of the redeposition processes on a perfectly planar surface, the contribution of which to the researched phenomena being characterized by the quantity I_z (see Eq. (15)).

The solution of Eq. (40) for the level of the equilibrium planar surface is trivial:

$$z_2 = \frac{YJ_2-\Lambda}{\lambda_1} = \frac{YJ_2-\Lambda}{\lambda-\alpha_0 I_z}. \tag{42}$$

The inequality $z_2 \geq 0$ and condition (41) result in the relationship

$$\Lambda \geq YJ_2. \tag{43}$$

The solution of Eq. (39) looks like

$$z_1(\rho) = h + \sqrt{R_0^2-\rho^2}. \tag{44}$$

It is easy to see that it describes the top hemisphere of radius R_0 and with the center located at the point $(\rho_0=0,\ h)$, where

$$R_0 = -\frac{YJ_2}{\lambda}, \quad h = -\frac{\Lambda}{\lambda}. \tag{45}$$

Solution (44) has to satisfy condition (38), so that the inequality

$$\lambda = \alpha_0 I_z + \lambda_1 < 0 \tag{46}$$

holds true.

From the requirement that the general solution (36) of Eq. (29) must be continuous, it follows that the solutions of Eqs. (39) and (40) have to match at the boundary $\rho=R$,

$$z_1(R) = z_2. \tag{47}$$

Whence, taking into account expression (44), the radius of the protrusion's base R (the critical dimension of an island) is determined as

$$R = \sqrt{R_0^2-(z_2-h)^2}, \tag{48}$$

where the quantities R_0, z_2, and h are determined in terms of the model parameters [see Eqs. (42) and (45)]. We point out that the continuity of solution (36) at the boundary $\rho=R$ is possible only if the inequality

$$h+R_0 \geq z_2 \geq h \tag{49}$$

is fulfilled.

Thus, within the scope of the assumptions made [see Eqs. (31)–(38)], the stationary profile of the surface looks like a solitary protrusion having the form of a spherical dome (a drop-shaped nucleus or island) against the background of the equilibrium planar surface at a level z_2. The radius of this dome's base (hence, its height too) is determined by formula (48). The equilibrium contact "wetting" angle ϕ is also determined by the model parameters and is given by the expression

$$\text{tg}\phi = -\left.\frac{dz}{d\rho}\right|_{\rho=R} = \frac{\sqrt{R_0^2-(z_2-h)^2}}{z_2-h}. \tag{50}$$

The first theoretical description of drop-shaped nuclei in the form of spherical domes was presented in work [9] (see also work [1]). In work [9], the authors substantially used the assumption about the anisotropy of the free energy of the interface if the linear tension effects are neglected. At the same time, the general assumptions of the model proposed by us allow the nuclei to be described in the form of Eqs. (36), (42), (44), and (47) (taking into account the sputtering of the surface), without taking advantage of explicit expressions for thermodynamic properties of the surface. We emphasize that, according to the model basics, the formation of such a relief is possible even in the case where the initial profile of the surface is planar and with a weakly non-uniform distribution of accommodation centers.

We also note that the assumption made about the solitude of a nucleus means, from the physical point of view, that the distance between two neighbor islands, which are being formed, is large in comparison with their linear dimensions.

5. Discussion of Results

The approach developed in this work is based, in essence, on the assumption that the evolution of the surface is defined at any moment by two characteristics: the surface profile and the distribution of accommodation centers over it. In the framework of this approach, the procedure of derivation of the equations describing the surface evolution and the general analysis of the solutions of those equations do not require one to know the explicit analytical forms for the thermodynamic properties of the surface. In particular, in the general case, the shape of the surface profile, which is determined by the solution of stationary Eq. (29) with regard for Eq. (35), can be different depending on the relationships between the model parameters (in this connection, see works [3, 4, 6, 7]). Among a lot of such relations, only conditions (38), (43), (46), (47), and (49) ensure that the "islands" should possess the shape of a spherical dome. In this case, condition (43) means that the contributions of the effects of deposition and adsorption to the process under investigation have an advantage over those made by the effects of sputtering of deposited particles by the ion beam.

However, in order to define the conditions for this or that solution of the evolution equations (22) to be realized, it is necessary, of course, to engage thermodynamic considerations. The role of these considerations is to specify the explicit forms for the model parameters and to determine their dependence on the thermodynamic characteristics of the surface (temperature, pressure, etc.). After that, there appears an opportunity to predict which type of the stationary solution of the equations describing the surface evolution can be feasible under the specific experimental conditions. From the above-said, it is clear that the proposed model allows one to classify the possible types of the surface relief that arises in the course of the deposition of particles onto the substrate surface, provided that an ion beam is bombarding the latter. The spherical-dome shape of nuclei or islands, which was described in the previous section, was selected only to demonstrate the efficiency of the approach developed in this work and its prospects for further researches.

It is evident that if the diffusion processes are neglected, the effects of sputtering play an essential role. It should be pointed out that, in the case of a negligibly low sputtering ($J_2 \to 0$, $T_z \to 0$), the linear dimensions of nuclei tend to zero, $R \to 0$ (see Eqs. (45) and (48)). This circumstance might have evidenced, at first glance, for the establishment of a stationary planar profile of the surface. However, judging from the obtained solutions (42) and (44), such a conclusion turns out to be incorrect. In this case, according to the initial points of the model, the conditions for the stationary (or quasistationary) modes of the surface evolution to exist are not satisfied. It is obvious that, in the case of a negligibly low sputtering, the surface diffusion must play the crucial role in the formation of the island-like relief of the surface.

The analysis presented in this work did not take the diffusion processes into account, although the consideration of the surface diffusion effects in the framework of our model does not cause basic difficulties (in this connection, see Eqs. (26) and (27). As was mentioned above, these effects were not accounted for, in the main, because of our desire to find analytical solutions of the obtained evolution equations. The account of the effects of surface diffusion and sputtering results in that the derived system of equations describing the surface evolution can be solved only numerically.

The experiments, which have been carried out by us in parallel with theoretical calculations, also aimed at studying the influence of the ion-beam irradiation on the nucleation mechanism for a film texture. The regularities in the formation of a Cromium coating were investigated at the stage when the film was not continuous yet, but comprised a set of isolated islands. Chromium was evaporated from an electron-beam evaporator either without the accompanying ion bombardment or under the irradiation with 30-keV nitrogen ions. The salt NaCl served as a substrate, the temperature was 200°C, the evaporation rate 0.15 nm/s, and the intensity of ion current 10^{14} ion/(cm^2 s). The research was carried out by using high-voltage electron microscopy. The results obtained have demonstrated an essential difference between the nucleation character of island structures in the cases with and without ion-beam irradiation. In the former case, the system of the nuclei of grains which are appeared has the distribution over size to be close to a Gaussian-like one. The peak is within a size interval of 2–3 nm. As the deposition duration increases, the shape of this curve does not change, but it shifts towards larger sizes. By the moment when the film becomes continuous, its structure consists of grains, the average size of which is equal to about 25–30 nm. A different situation arises, when Cromium is deposited under conditions of the bombardment with nitrogen ions. By the moment when the average size of islands reaches 2–3 nm, their distribution over sizes is close to that obtained in the case where a film is deposited without accompanying ion irradiation. But afterwards, the picture changes substantially. The areas with islands 6–8 nm in size appear in the microscope field of view. In the vicinity of these islands, there are the areas depleted Cromium nuclei. We interpret this effect as the coalescence of small grains owing to the surface diffusion of Cromium. The following stage of the process is the formation of a new generation of islands around large grains that have been formed due to coalescence. In the final state of the film, when it is a continuous object, the grain distribution over sizes has two peaks within the ranges of 2–3 and 8–10 nm. Thus, the experiment showed that the bombardment with nitrogen ions, which accompanies the process of Cromium deposition, stimulates the surface diffusion, on the one hand, and enhances the generation of new accommodation centers for the grains to nucleate, on the other hand. This fact is in agreement with the theoretical calculations of this work.

At last, we recall that, in the previous section, the equations of evolution were named “stationary”for convenience. The analysis of the description of the process at the stage, when the nuclei of coverings are being formed on the substrate surface, shows that the relevant solutions should be, most likely, quasistationary; this is really observed in experiments. In this connection, there arises a question concerning the stability of the “stationary” solutions. The study of the stability of the “stationary” solutions of the evolution equations obtained for the surface profile remains the subject of our present interest.

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КІНЕТИКА ФОРМУВАННЯ ТОНКОПЛІВКОВИХ ПОКРИТТІВ В УМОВАХ БОМБАРДУВАННЯ ВАЖКИМИ ІОНАМИ

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Р е з ю м е

Запропоновано модель опису початкової стадії зародження та еволюції тонкоплівкових покриттів в умовах бомбардування поверхні пучком іонів. Одержано рівняння еволюції профілю поверхні та функції розподілу центрів адсорбції. Знайдено квазістаціонарні розв'язки одержаних рівнянь у випадку слабконеоднорідного розподілу центрів адсорбції та плаского профілю поверхні у початковий момент часу. У термінах параметрів моделі визначено умови формування зародків у вигляді сферичного купола.