LOW-DIMENSIONAL SYSTEMS
AND SURFACE PHYSICS

Self-Organization of Plasma–Condensate Quasi-Equilibrium Systems

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Received July 9, 2008

Abstract—The steady-state regime of condensation under conditions close to the phase equilibrium is shown to be provided by the self-organization of the plasma–condensate system. This self-organization is associated with the fact that the presence of the plasma leads to a considerable increase in the energy of adatoms and, hence, in the temperature of the growth surface in the course of their condensation. The increase in the temperature is compensated by the desorption flow of adatoms responsible for the supersaturation. It is demonstrated that the proximity of the system to the phase equilibrium ensures the selective regime of condensation during which the adatoms are attached onto active centers of the growth surface, thus forming various three-dimensional structures.

PACS numbers: 05.65.+b, 68.55.-a, 81.15.Aa

DOI: 10.1134/S1063783409050266

1. INTRODUCTION

The development of nanotechnologies associated with the condensation of matter comes to the solution of two main problems: the maintenance of the steady-state regime of condensation, on the one hand, and the creation of conditions close to the phase equilibrium, on the other hand [1–4]. Owing to the sensitivity of quasi-equilibrium condensation to the choice of technological parameters, the former problem is solved by providing a constant supersaturation. At low supersaturations, the system is insignificantly far from equilibrium, close to which the condensation occurs in a self-organized manner, so that only the adatoms forming the strongest chemical bonds are fixed on the growth surface. As a result, the thermodynamic potential of the system acquires a minimum value, as is the case with the energy of elastic stresses at the condensate–substrate interface [5].

At present, the technological solution to the above problems has been implemented using the molecular-beam epitaxy and the condensation conditioned by the interaction of reactive media with the growth surface (chemical vapor deposition techniques, condensation upon decomposition of organometallic compounds, crystallization in weakly supersaturated solutions, electrolytic deposition). In this case, the deviations of the temperature ΔT and the pressure ΔP from the equilibrium values of the temperature T of the growth surface and the pressure P of the molecular flow lead to the difference between the chemical potentials $\Delta\mu \equiv \mu_c - \mu_v$ of the condensate and the vapor of the deposited material [6, 7]; that is,

$$\Delta\mu = q\frac{\Delta T}{T} - T\frac{\Delta P}{P}, \qquad (1)$$

where q is the heat of evaporation of an atom (here, the atomic volume of the condensate is ignored and the vapor is assumed to be an ideal gas). According to relationship (1), the conditions close to the phase equilibrium are relatively easily created in volatile materials which, even at relatively low temperatures of the growth surface, have high saturation vapor pressures. These conditions substantially limit the capabilities of molecular-beam epitaxy. In this respect, it is more preferable to use chemical methods in which the equilibrium state is achieved not only as the result of an increase in the temperature and a weakening of flows of deposited materials but also due to the desorption of atoms during back chemical reactions.

The providing of the self-organization in all the aforementioned technologies requires a stringent control over the conditions of the steady-state condensation process. Therefore, we are based on the scheme according to which the self-organization is ensured by the natural course of the condensation process, so that its quasi-equilibrium state is achieved by the presence of the plasma increasing the effective temperature of the growth surface [1–4].

From the microscopic viewpoint, the action of the low-temperature plasma that brings the plasma–condensate system closer to equilibrium is determined by
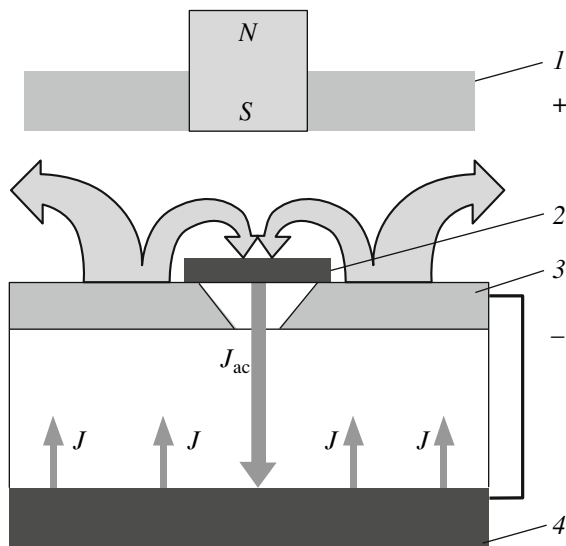
Fig. 1. Block diagram of the self-organized sputtering system: (*1*) anode with the magnet, (*2*) damper, (*3*) cathode to be sputtered, and (*4*) substrate.

Fig. 2. Distributions of (a) flows and (b) concentrations in the surface layer.

the fact that the plasma particles collide with the growth surface and favor the evaporation and sputtering of weakly bound adatoms. An example of this action is the process of homonucleation of nanotubes and fullerenes in an arc discharge, where the influence of the plasma favors the breaking of all chemical bonds, except for the strongest sp^2-hybridized bonds. One more example is provided by the formation of diamond-like nanostructures under the action of plasma flows [1]. It is characteristic that both processes involve the condensation of carbon for which the low volatility is compensated by the action of the plasma increasing the effective temperature of the growth temperature. However, the arc discharge does not provide the steady-state course of the process and hinders the growth of monocrystals and the formation of statistically homogeneous island and porous structures.

The aim of this study is to elucidate the mechanism of the self-organization process underlying the experimental technique developed in our earlier works [1–4]. In Section 2, we show that the sputtering system involving the anode with a built-in magnet and the hollow cathode with a substrate located in its inner part makes it possible to produce a concentrated plasma (in the vicinity of the growth surface) that provides the steady-state condensation of adatoms characterized by a low supersaturation. In Section 3, we develop the synergetic scheme according to which the presence of the plasma leads to an increase in the energy of adatoms so that their condensation increases the temperature of the growth surface. The increase in the temperature of the growth surface is compensated by the desorption of adatoms. In the framework of this scheme, the kinetic representation of the self-organization of flows is
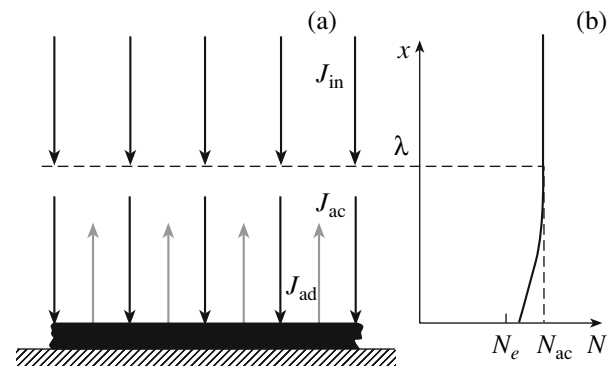
described in Subsection 3.1 and the thermodynamic representation explaining the concentration distribution of the material to be sputtered is described in Subsection 3.2. Finally, in Section 4, we demonstrate that, owing to the proximity of the system to the phase equilibrium, only adatoms forming the strongest bonds are fixed on the growth surface. This results in the selective regime of condensation, during which the adatoms are built at active centers of the growth surface.

2. PROVIDING OF SELF-ORGANIZATION IN A SPUTTERING SYSTEM

The self-organized regime of quasi-equilibrium steady-state condensation has been implemented in ion-plasma systems. The block diagram of these systems is shown in Fig. 1. One of the main elements of the self-organized sputtering system is an anode with a built-in magnet. The action of this magnet favors a multiple increase in the density of the plasma in an inert-gas medium. One more fundamentally important component is a hollow cathode, which in its inner part contains the substrate separated from the anode by an inlet hole and a damper (this design allows one to produce a highly concentrated plasma consisting of ions of the sputtered material and the inert gas in the vicinity of the growth surface). First, high vacuum (with a residual pressure of 10^{-8} Pa) is produced in a chamber. Then, the filling of the chamber with the inert gas to a pressure of 20–40 Pa and its purifying are performed (this high pressure reduces the mean free path to several micrometers). The target is sputtered under the action of a glow discharge, and the steady-state course of this process is provided by the magnetron effect and the cumulative effect of the hollow cathode.

For the closed damper, the steady-state regime is ensured by the fact that the sputtered material with the flow determined by the discharge power is completely condensed on adjacent parts of the vacuum chamber. After the inlet hole was opened, the electric field pulls positive ions in the inner part of the cathode, which ini-

tially contains the inert gas. Owing to the relatively small volume (10–380 cm^3), the ion drift leads to a rapid increase in the pressure in the accumulation system. In this case, a variation in the total concentration N_{ac} of atoms and ions of the sputtered material inside the cathode is defined by the equation

$$\dot{N}_{ac} = L^{-1}(J_f - J_P) + l^{-1}(J_{in} - J_{ad}). \qquad (2)$$

Hereinafter, the dot over symbols will mean the differentiation with respect to the time t. In expression (2), the drift flows J_f and J_P are determined by the change in the electric field and the pressure difference at characteristic lengths L, respectively, and the diffusion flows J_{in} and J_{ad} acting at distances of the order of l are determined by the inflow through the inlet hole and the adsorption on the substrate. Owing to the limited volume of the accumulation system, the first two flows very rapidly compensate for each other ($J_f = J_P$) and a further variation in the concentration N_{ac} is determined by the interdiffusion of the inert gas and the sputtered material due to the inlet flow J_{in} and the adsorption flow J_{ad}. Irrespective of the external flow in the absence of adsorption ($J_{ad} = 0$), the diffusion process results in equalization of the concentrations at the inlet of the hollow cathode and inside it. As a consequence, the accumulation flow becomes equal to zero ($J_{in} = 0$). The steady-state adsorption process, during which the concentration of the sputtered material in the accumulation system remains constant ($\dot{N}_{ac} = 0$), is provided under the condition that the diffusion flows associated with the inflow through the inlet hole and the adsorption on the substrate compensate for each other; that is,

$$J_{in} = J_{ad}. \qquad (3)$$

The adsorption of ions of the sputtered material on the growth surface is insured by processes proceeding in the cathode space of the glow discharge. The thickness of the near-cathode layer, in which the potential predominantly drops, is determined by the Debye screening length λ. The Debye screening length and the diffusion coefficient are represented by the equalities [6, 8]

$$\lambda^2 = \frac{\varepsilon T_p}{4\pi e^2 N_i}, \quad D = \frac{\sigma T_p}{e^2 N_i}. \qquad (4)$$

Here, ε is the dielectric permeability of the plasma; σ is the conductivity of the plasma; T_p is its temperature measured in energy units; and e and N_i are the charge and the total concentration of ions of the deposited material and the inert gas in the accumulation system, respectively. The atomic concentration of the sputtered material at the upper boundary of the near-cathode layer is equal to the accumulated concentration N_{ac} defined by Eq. (2). The lower boundary of this layer is the growth surface, at which the volume concentration of atoms is N (under quasi-equilibrium conditions, this

concentration $N \geq N_e$ insignificantly exceeds the equilibrium concentration N_e). Owing to condition (3), the adsorbed flow is reduced to the diffusion component determined by the Onsager relation

$$J_{ad} \equiv D|\nabla N| \simeq \frac{D}{\lambda}(N_{ac} - N). \qquad (5)$$

At the beginning of the condensation, when the conditions are equilibrium, this flow can be written in the form

$$J_{ac} = \frac{D}{\lambda}(N_{ac} - N_e), \qquad (6)$$

which insignificantly exceeds the steady-state flow (expression (5)); that is,

$$J_{ac} - J_{ad} \ll J_{ac}. \qquad (7)$$

The quasi-equilibrium condition

$$N - N_e \ll N_e \qquad (8)$$

is provided by the highly concentrated plasma. The presence of this plasma increases the substrate temperature, so that the plasma brings not only materials with a high volatility but also metals and semiconductors (in which it is very low) closer to the phase equilibrium.

As can be seen from Fig. 2, the inflow of the deposited material is provided by the diffusion component (relationship (6)), which is compensated by the counter flow of adatoms (expression (5)) evaporated from the substrate under the influence of the plasma. In this case, the structure of the near-cathode layer is determined by the adsorption of ions and the desorption of neutral atoms of the deposited material. The collisions with plasma particles lead to the ionization of these atoms, and the intensity of this process increases with an increase in the pressure of the working gas. As a result, with distance from the substrate, the volume concentration of ions increases from the minimum value N to the maximum concentration N_{ac}, which is observed at the characteristic distance λ determining the width of the near-surface layer. This variation in the concentration ensures the diffusion flow (formula (5)).

3. SELF-ORGANIZATION OF THE PLASMA– CONDENSATE SYSTEM

Since the self-organization is determined by the processes occurring on the growth surface, it is necessary to change over from the volume concentration N to the surface concentration $n \equiv Na$, where the scale factor a is defined by equality (17). As is known, the self-organization process can be investigated in the framework of a pair of conjugated concepts [9]. Subsection 3.1 is devoted to the study of the self-organization process by analyzing the flows, and the corresponding concentration distribution is examined in Subsection 3.2.

3.1. Self-Organization of Flows

The self-organization process is determined by the supersaturation $n - n_e$ of adatoms on the growth surface, where its decrease is ensured by the desorption flow J and an increase is provided by the diffusion component J_{ad} associated with the adsorption flow (relationship (5)).[1] In the presence of the condensate when all atoms evaporate from the substrate, the former component meets the condition $J = -J_{ac}$, where the accumulated flow J_{ad} is defined by equality (6). For the diffusion component, the continuity equation $\dot{n}/a + \nabla\mathbf{J}_{ac} = 0$, in which the action of the source is estimated as $|\nabla\mathbf{J}_{ad}| \simeq J_{ad}/\lambda \simeq (D/\lambda^2)(n - n_e)/a$, results in $\dot{n} \simeq -(D/\lambda^2)(n - n_e)$. The variation in the concentration $n(t)$ under the action of both flow components is described by the equation

$$\dot{n} = \frac{n_e - n}{\tau} - J. \tag{9}$$

Here, the relaxation time is defined by the equalities

$$\tau \equiv \frac{\lambda^2}{D} = \frac{\varepsilon}{4\pi\sigma}, \tag{10}$$

where the former equality includes relationships (4). In the steady-state regime, when $\dot{n} = 0$ and the self-organization is absent ($J = -J_{ac}$), the supersaturation

$$n_{ac} - n_e = \tau J_{ac} \tag{11}$$

determined by the accumulation flow (expression (3)) is attained for the relaxation time. According to Eq. (9), with a deviation from the steady state, the supersaturation defined by expression (11) is achieved as a result of the Debye relaxation with the time determined by relationship (10).

The initiation of the self-organization process is associated with the following circumstances. As the supersaturation $n - n_e$ increases, the condensed adatoms transfer an excess of their energy to the growth surface and increase its temperature T. This enhances the evaporation of atoms of the deposited material, which is ensured by the desorption flow J compensating for the initial supersaturation.[2] The representation of the sequential mechanism of the quasi-equilibrium condensation process requires the self-consistent description of the time dependences of the adatom concentration $n(t)$, the growth surface temperature $T(t)$, and the desorption flow $J(t)$. In the framework of the synergetic approach, the evolution equations contain dissipative contributions of the above quantities and the terms representing the positive and negative feedbacks, so that

their balance provides the self-organization process [10]. In particular, in Eq. (9), the first term is the dissipation contribution and the second term describes the linear relation between the rate of change in the concentration and the desorption flow. The variation in the growth surface temperature is described in the same way

$$\tau_T \dot{T} = -T - a_T nJ, \tag{12}$$

where τ_T is the corresponding relaxation time and $a_T > 0$ is the coupling constant. Unlike Eq. (9), here, it is assumed that the dissipation results in the relaxation of the temperature to zero value rather than to a finite value and the last term represents a nonlinear relation between the concentration and the flow, which decreases the rate of change in the temperature. In order to ensure the self-organization, the negative contributions in relationships (9) and (12) should be compensated by the positive component in the evolution equation of the flow; that is,

$$\tau_J \dot{J} = -(J_{ac} + J) + a_J nT. \tag{13}$$

Here, τ_J is the time of relaxation to the opposite value of the accumulation flow $-J_{ac}$ and $a_J > 0$ is the coupling constant that provides an increase in the flow due to the mutual effect of the adatom concentration and the growth surface temperature.

Equations (9), (12), and (13) form the synergetic Lorenz system, within which the supersaturation $n - n_e$ is reduced to the order parameter, the growth surface temperature plays the role of the conjugate field, and the desorption flow J is the control parameter [10]. In the dynamic equilibrium state, when the dependence on the time is absent, Eq. (9) leads to the constant supersaturation (expression (11)) determined by the accumulation flow (formula (3)). Correspondingly, equality (12) results in the dependence $T = a_T n|J|$, according to which the growth surface temperature increases as a result of a mutual increase in the concentration of adatoms and the flow of their desorption. Finally, Eq. (13) determines the steady-state flow $J = -J_{ac} + a_J nT$, which increases from the value $-J_{ac}$ determined by the accumulation system due to the simultaneous increase in the adatom concentration and the temperature.

As a rule, in the course of condensation, the following adiabatic conditions are satisfied:

$$\tau_T \tau_J \ll \tau, \tag{14}$$

which imply that, during the evolution of the system, the temperature $T(t)$ and the flow $J(t)$ follow the variation in the adatom concentration $n(t)$. As a consequence, the time derivatives can be disregarded in Eqs. (12) and (13), which lead to the simple relationships

$$T = a_T J_{ac} \frac{n}{1 + a^4 n^2}, \tag{15}$$

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[1] Since the desorption flow $\mathbf{J}$ is directed from the growth surface, its magnitude satisfies the condition $J < 0$ and the magnitude of the adsorption flow obeys the inequality $J_{ad} > 0$.

[2] The increase in the substrate temperature T also enhances the evaporation of atoms of the inert gas; however, this only insignificantly changes the plasma temperature T_p and does not affect the flow J.
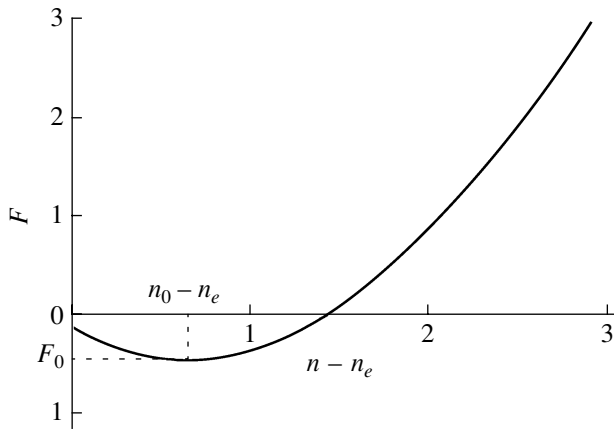
Fig. 3. Dependence of the effective potential on the supersaturation at $a = 1$, $n_e = 0.1$, and $\tau J_{ac} = 1$.
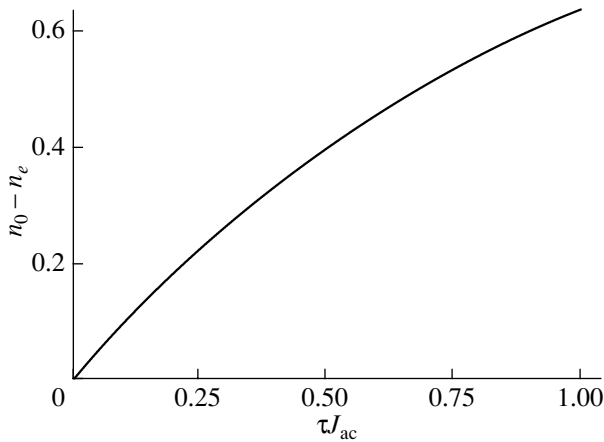

Fig. 4. Steady-state supersaturation as a function of the accumulated flow J_{ac} and the relaxation time τ at $a = 1$ and $n_e = 0.1$.
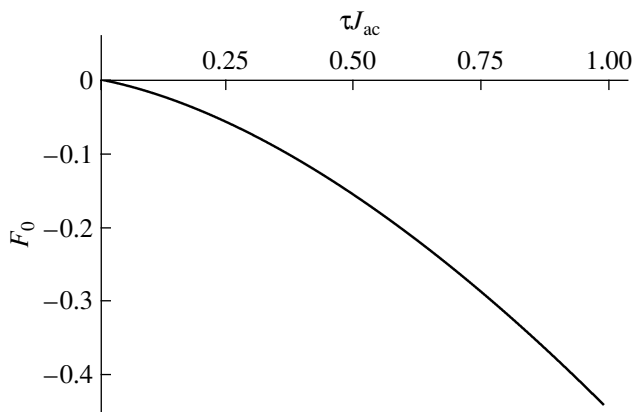

Fig. 5. Steady-state effective potential as a function of the accumulated flow J_{ac} and the relaxation time τ at $a = 1$ and $n_e = 0.1$.

$$J = \frac{-J_{ac}}{1 + a^4 n^2}, \qquad (16)$$

where we introduced the characteristic size

$$a \equiv (a_T a_J)^{1/4}. \qquad (17)$$

Equalities (15) and (16) demonstrate that, as the adatom concentration increases, the growth surface temperature initially linearly increases and then, at $n = a^{-1}$, reaches saturation corresponding to the maximum temperature $T_{max} = \sqrt{a_T/a_J}\,J_{ac}/2$. In this case, the desorption flow increases from $J_{min} = -J_{ac}$ to $J_{max} = -J_{ac}/2$.

Substitution of relationship (16) into Eq. (9) shows that the adiabatic evolution of the system is described by the Landau–Khalatnikov equation

$$\tau \frac{\partial n}{\partial t} = -\frac{\partial F}{\partial n}, \qquad (18)$$

in which the effective potential has the form

$$F = \left(\frac{n^2}{2} - n_e n\right) - \frac{\tau J_{ac}}{a^2} \arctan(a^2 n). \qquad (19)$$

As can be seen from Fig. 3, the minimum value F_0 of this potential is observed at the steady-state point n_0 with the position determined by the equation

$$(n_0 - n_e)(1 + a^4 n_0^2) = \tau J_{ac}. \qquad (20)$$

The solution to this equation is presented in Fig. 4. It can be seen from this figure that, as the accumulated flow J_{ac} and the relaxation time τ increase, the steady-state supersaturation n_0 first linearly increases from the equilibrium value n_e and then this increase is retarded. Since the supersaturation is considerably smaller than the equilibrium value ($N_0 - n_e \ll n_e$), the concentration in the last factor of Eq. (20) can be taken equal to the equilibrium concentration. As a result, the steady-state supersaturation takes the form

$$n_0 - n_e \le \frac{\tau J_{ac}}{1 + a^4 n_e^2} \le \tau J_{ac}. \qquad (21)$$

As can be seen from Fig. 5, the steady-state value of the effective potential (expression (19)) decreases monotonically with an increase in the accumulated flow J_{ac}, the relaxation time τ, and the equilibrium concentration n_e. This behavior is associated with the estimate

$$F_0 \simeq -\frac{n_0^2}{2} \simeq -\frac{1}{2}(n_e + \tau J_{ac})^2. \qquad (22)$$

It is characteristic that the steady-state Eq. (9) for the flow $J = -J_{ac}$ leads to the last relationship in conditions (21), which is reduced to expression (11). In turn, this expression differs from the penultimate equality in conditions (21) by the absence of the denominator determined by the equilibrium concentration n_e and the scale

a. This denominator describes the effect of self-organization, which, according to the Le Chatelier's principle, suppresses the direct action of the flow. However, this factor has no radical effect due to the smallness of the quantity τJ_{ac}. Actually, at large values of the quantity τJ_{ac}, the dependence $n_0(n_e)$ following from the penultimate relationship in conditions (21) should acquire a nonmonotonic character, whereas the initial Eq. (20) always possess the only root.

According to equalities (15) and (16) and the last estimate in conditions (21), in the steady state, the magnitude of the desorption flow $|J_0|$ and the growth surface temperature T_0 are written in the form

$$|J_0| = \frac{n_0 - n_e}{\tau} \le J_{ac},$$

$$T_0 = \frac{a_T}{\tau} n_0(n_0 - n_e) \le a_T n_e J_{ac}. \tag{23}$$

This leads to the important inference: the self-organized condensation process occurs so that the desorption flow J_0 almost compensates for the accumulated flow J_{ac} and weakly affects the initial temperature of the growth surface $T_{ac} = a_T n_e J_{ac}$. Since the difference

$$J_c \equiv J_{ac} + J_0 = J_{ac} - |J_0| \tag{24}$$

ensures the inflow of the sputtered materials to the condensate, the smallness of this difference ($J_c \ll J_{ac}$) reflects the quasi-equilibrium character of the self-organization process. However, estimates (21) and (22) demonstrate that the steady-state quantities n_0, $|J_0|$, and T_0 are determined by the accumulation flow J_{ac} in the volume of the hollow cathode rather than by the sputtered target located outside the cathode. Since the quantity J_{ac} depends only on the design of the sputtering system, the steady-state regime of the condensation process is retained irrespective of the sputterer operation.

3.2. Self-Organization of Concentrations

The above mechanism of self-organization is based on the fact that the condensation of adatoms having an increased energy leads to an increase in the growth surface temperature T and the desorption flow J of adatoms responsible for the supersaturation compensates for this increase. According to formula (24), only a small portion J_c of the adsorbed flow J_{ac} is expended for the inflow of condensed adatoms and its main component is compensated by the desorption flow J. This separation is conveniently represented in the form of annular flows (Fig. 6a) with the intensity determined by the desorption component J. It can be seen from Fig. 6b that the annular flows do not make contributions to the condensation, because the ions forming these flows are accelerated by the electric field so strongly that their energy becomes sufficient for intensive migration over the substrate surface, during which the adatoms initially evaporate and then are ionized under the action of
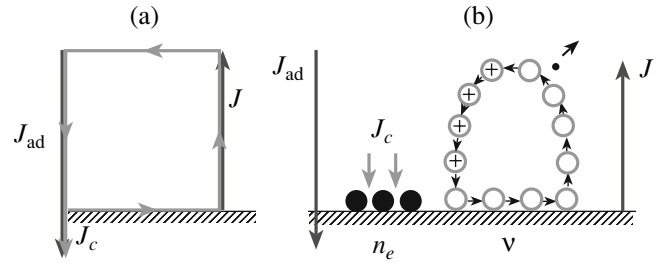

Fig. 6. (a) Formation of annular flows in the vicinity of the growth surface and (b) separation of atoms into the condensate n_e and the supersaturation component ν.

colliding plasma particles. As a result, the self-organization process is determined by the concentration of annular flows, so that an increase in this concentration compensates for the increase in the growth surface temperature due to the condensation of adatoms. Since the concentration ν of annular flows increases monotonically with the supersaturation of adatoms, this concentration can be used instead of the desorption flow J. This ensures the changeover from the aforementioned kinetic representation to the thermodynamic representation [9].

With due regard for the conventional synergetic scheme [10], let us consider a self-consistent variation in the concentration of adatoms n and annular flows ν with a variation in the growth surface temperature T. The time dependences of these quantities are determined by the system of equations

$$\tau \dot{n} = (n_e - n) + \nu, \tag{25}$$

$$\tau_T \dot{T} = -T + b_T n \nu, \tag{26}$$

$$\tau_\nu \dot{\nu} = (\nu_{ac} - \nu) - b_\nu n T, \tag{27}$$

which are reduced to the system of equations (9), (12), and (13) upon replacement ν by $\tau|J|$ (moreover, here, the feedback constants b_T and b_ν are renormalized). Equalities (6) and (10) reflect the accumulated content of annular flows; that is,

$$\nu_{ac} \equiv \tau J_{ac} = \lambda(N_{ac} - N_e), \tag{28}$$

which determines the behavior of the system. Under the adiabatic conditions τ_T and $\tau_\nu \ll \tau$, Eqs. (26) and (27) lead to relationships of type (15) and (16); that is,

$$T = b_T \nu_{ac} \frac{n}{1 + b^4 n^2}, \quad \nu = \frac{\nu_{ac}}{1 + b^4 n^2} \tag{29}$$

with the characteristic scale $b \equiv (b_T b_\nu)^{1/4}$. As the adatom concentration n increases from zero to the limiting value b^{-2}, the condensate temperature T reaches the maximum value $T_{max} = \sqrt{b_T/b_\nu}\,\nu_{ac}/2$ and the concentration ν of annular flows decreases from the accumulated value ν_{ac} to $\nu_{ac}/2$. Substitution of the last equality (29)
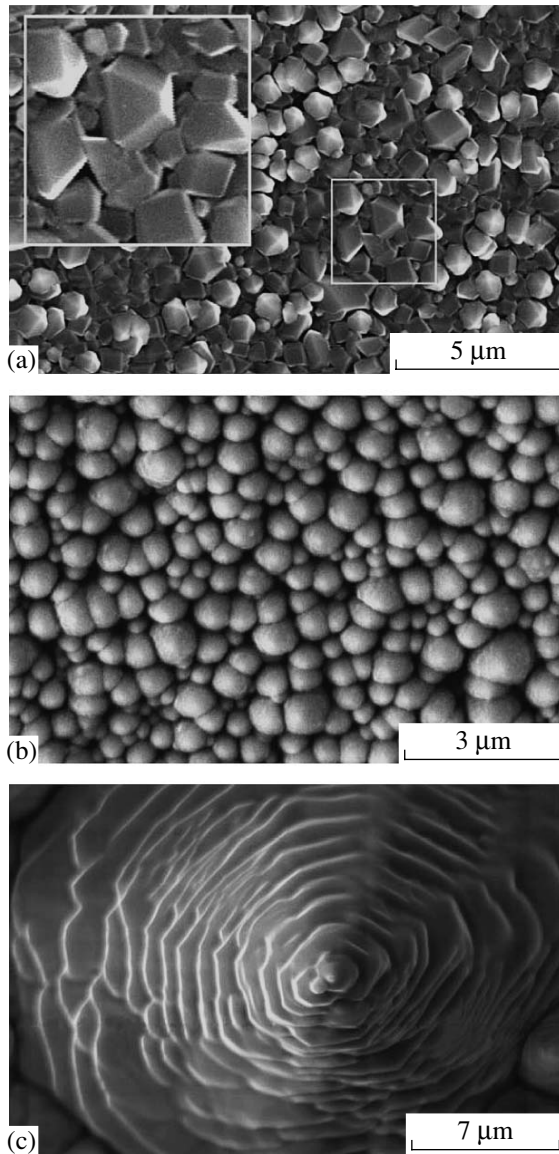
Fig. 7. Examples of structures prepared through quasi-equilibrium steady-state condensation: (a) structure of aluminum layers with the (210) habit plane of microcrystals, (b) structure of copper layers with an atomically rough surface, and (c) nickel single crystal grown under conditions of field selectivity.

into expression (25) results in the Landau–Khlatnikov equation (18) with the effective potential (relationship (19)), where the accumulated concentration of annular flows (formula (28)) should be used instead of the quantity τJ_{ac}. As a result, we obtain the relationships

$$\nu_0 = n_0 - n_e \le \frac{\nu_{ac}}{1 + a^4 n_e^2} \le \lambda\left(N_{ac} - \frac{n_e}{a}\right). \qquad (30)$$

The first relationship demonstrates that the concentration of annular flows is reduced to the supersaturation of adatoms with the steady-state value determined by

the accumulated concentration N_{ac} of ions of the sputtered material with respect to the equilibrium value n_e/a. According to expressions (4), an increase in the total concentration N_i of ions in the accumulation system leads to a decrease in the Debye screening length λ, which, in turn, results in a decrease in the steady-state supersaturation (expression (30)).

4. DISCUSSION

The performed investigation shows that the self-organization of the plasma–condensate system ensures the steady-state regime of condensation under weak-supersaturation conditions close to the phase equilibrium. This self-organization is associated with the fact that the presence of the plasma leads to an increase in the adatom energy; as a result, their condensation favors an increase in the temperature of the growth surface. The increase in the temperature is compensated by the desorption flow of adatoms responsible for the supersaturation. In this case, only a small portion of the adsorbed flow is expended for the inflow of condensed adatoms and its main component is compensated by the desorption flow. This behavior is explained by the presence of annular flows that do not make contributions to the condensation, because the ions forming these flows intensively migrate over the substrate surface and then evaporate under the action of colliding plasma particles. As a result, the self-organization process is determined by the concentration of annular flows, so that an increase in this concentration compensates for the increase in the growth surface temperature upon condensation of adatoms.

Owing to the proximity of the system to the phase equilibrium due to the self-organization process, only adatoms forming the strongest bonds are fixed on the growth surface. This results in the selective regime of condensation, during which the adatoms are built at active centers of the growth surface, such as kinks of crystal growth monosteps, crystal lattice defects, intergrown grain boundaries, etc. It can be seen from the scanning electron microscopy image of the aluminum condensate (Fig. 7a) that, at distances of less than 5 μm, the crystallization pattern is determined by the layer-by-layer growth of the (210) crystallographic plane on the first of the above centers. However, a large-scale pattern is associated with the leading growth of the condensate on different crystallization centers with the formation of a mosaic structure. The contribution of these centers most clearly manifests itself upon changing over from the layer-by-layer crystal growth to the normal growth observed on the atomically rough surface (Fig. 7b). Finally, at low working gas pressures and high voltages at the anode, the condensation pattern is determined by field selectivity, which provides the condensation of adatoms in growth surface regions with a maximum curvature (Fig. 7c).

The examples shown in Fig. 7 demonstrate that the self-organization of the plasma–condensate system ensures the steady-state regime of condensation, during which the adatoms are built at active crystallization centers with the formation of structures characterized by different architectures. The sequence of formation of these structures is determined by the energy of bonding between the adatoms and the growth surface. The analysis of the data obtained in our earlier works [1–4] allows us to draw the conclusion that, as the bonding energy increases, the contribution of different crystallization centers changes in the following order: (1) the layer-by-layer crystallization on kinks of crystal growth monosteps, (2) the normal crystal growth on atomically rough surface, (3) the leading growth of the condensate on different crystallization centers, and (4) the condensation under conditions of field selectivity.

ACKNOWLEDGMENTS

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Translated by O. Borovik-Romanova