

Description of the Stochastic Condensation Process under Quasi-Equilibrium Conditions

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The system of three differential equations describing the stochastic condensation process under quasi-equilibrium conditions is constructed taking into account the additive and multiplicative components. The phase diagram of the system states was constructed and analyzed. The domains of the existence of the condensation processes, disassembly of previously deposited material, and the complete evaporation were determined. The distribution density of the concentration of adsorbed atoms was defined.

Keywords: Condensation, Disassembly, Self-Organization, the Fokker-Planck Equation.

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1. INTRODUCTION

Recently, the development of various branches of science and technology closely connected with the use of nanomaterials. At the same time the use of nanoporous materials, which are widely used in biology and medicine, is of particular interest. One of the promising directions for the production of such materials is a condensation process in the steady-state regime close to the phase equilibrium. This state is achieved due to the presence of the plasma, which increases the effective temperature of the growing surface. Thus the self-organization is provided by the natural course of the condensation process [1-3]. From a physical point of view, this self-organization is caused by the fact, that under the influence of the plasma the increasing of the adsorbed atoms energy leads to the increase of the growing surface temperature. The last is compensated by the flow of desorption of the adsorbed atoms responsible for the supersaturation $n-n_e$.

As a result of such processes in the systems with a weak feedback for small values of the equilibrium concentration n_e and deposited flux the only state, that meets the stationary regime of condensation, is realized. With increased feedback and inversion of the deposited flow a steady-state disassembly of the previously deposited condensate appears. This state corresponds to the mode of the nanoporous structures formation. The theoretical consideration of this process [4, 5] presents a self-consistent description of the three degrees of freedom: the surface concentration of adsorbed atoms n , the temperature T of the growing surface, measured from the ambient temperature, and the desorption flow J .

2. BASIC EQUATIONS

Using the previous example [4, 5], the representation of the self-organizing system reduces to three evolution equations for the corresponding degrees of freedom: the order parameter n , the conjugate field T , and the control parameter J .

The evolution equation for the surface concentration of adsorbed atoms is determined by the continuity condition and the Gauss theorem. The equations for the

remaining degrees of freedom contain dissipative terms and the contributions that represent the positive and negative feedbacks. In addition we take into account the stochastic sources, reflecting the random influence of the external factors, which always takes place in a real experiment. In this case for the temperature we consider the case of additive noise, and for the flow – multiplicative. This is because the temperature fluctuations depend only on the external conditions (e.g., the stability of the cooling device), and fluctuation of the flow of desorption is always linked to the surface concentration of adsorbed atoms.

As a result, the system of equations describing the stochastic process of condensation takes the form

$$\begin{aligned}\tau_n \dot{n} &= (n_e - n) - \tau_n J, \\ \tau_T \dot{T} &= -T - a_T n J + \zeta(t), \\ \tau_J \dot{J} &= -(J_{ac} + J) + a_J n T + n \xi(t),\end{aligned}\quad (2.1)$$

where τ_n , τ_T , τ_J are the relaxation times of the corresponding quantities, $a_T a_J$ – the positive coupling constants, and J_{ac} is an accumulated flow. The stochastic sources are represented as Ornstein-Uhlenbeck process

$$\begin{aligned}\langle \zeta(t) \rangle &= 0, \quad \langle \zeta(t) \zeta(t') \rangle = \frac{I_T}{\tau_\zeta} \exp\left(-\frac{|t-t'|}{\tau_\zeta}\right), \\ \langle \xi(t) \rangle &= 0, \quad \langle \xi(t) \xi(t') \rangle = \frac{I}{\tau_\xi} \exp\left(-\frac{|t-t'|}{\tau_\xi}\right).\end{aligned}\quad (2.2)$$

Here I_T , I are the intensities of the temperature and flow fluctuations, τ_ζ , τ_ξ – corresponding relaxation times.

For the analysis of this system is convenient to use dimensionless variables, relating the time t , concentration of adsorbed atoms n , the temperature T of the growing surface, the desorption flow J , and the fluctuations intensities I_T , I to the scales

$$t_s \equiv \tau_n, \quad n_s \equiv (a_T a_J)^{-1/2}, \quad T_s \equiv (\tau_n a_J)^{-1}, \quad J_s \equiv \tau_n^{-1} (a_T a_J)^{-1/2}, \quad I_s \equiv \tau_n.$$

Due to the coherence in the fluctuation processes the time of changes in the temperature and flow fluctuation

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tuations will be closely interrelated, thus it is possible to provide an approximation $\tau_c \approx \tau_\xi = \tau$. In addition, to facilitate the mathematical calculations it is appropriate to use the following relation for the intensity of noise $I_T = k_2 I$ where the coefficient k^2 shows how much the intensity of temperature fluctuations more than the intensity of flow fluctuations.

As a result, the dimensionless system of equations describing the stochastic system of plasma-condensate takes the form

$$\begin{aligned} \dot{n} &= -(n - n_e) - J, \\ \delta^{-1} \dot{T} &= -T - nJ + \kappa \lambda(t), \\ \sigma^{-1} \dot{J} &= -(J_{ac} + J) + nT + n\lambda(t), \end{aligned} \quad (2.3)$$

where $\delta^{-1} \equiv \tau_T / \tau_n$, $\sigma^{-1} \equiv \tau_J / \tau_n$, $\kappa = a_j k$ and

$$\langle \lambda(t) \rangle = 0, \quad \langle \lambda(t) \lambda(t') \rangle = \frac{I}{\tau} \exp\left(-\frac{|t-t'|}{\tau}\right) \quad (2.4)$$

It is known [6], that in general the system (2.3) does not have an analytical solution, so we will take into account the approximation $\tau_J \approx \tau_n \gg \tau_T$ ($\delta^{-1} \rightarrow 0$, $\sigma \approx 1$) Then the expression on the left side of the second equation of system (2.3) is negligibly small. Finally, instead of the system of three differential equations, we arrive at a motion equation for the nonlinear stochastic oscillator

$$\ddot{n} + \dot{n}\gamma(n) = f(n) + g(n)\lambda(t) \quad (2.5)$$

Where the friction coefficient $\gamma(n)$, the force $f(n)$ and the amplitude of the noise $g(n)$ are given by the expressions

$$\begin{aligned} \gamma(n) &= 1 + \sigma(1 + n^2), \\ f(n) &= \sigma [J_{ac} - (n - n_e)(1 + n^2)], \\ g(n) &= \sigma(1 + \kappa)n. \end{aligned} \quad (2.6)$$

3. THE STATIONARY SOLUTION OF THE FOKKER-PLANCK EQUATION

Based on the method, first proposed in [7] and developed in paper [8], the motion equation (2.4) can be associated with Fokker-Planck equation [9]

$$\frac{\partial P(n,t)}{\partial t} = -\frac{\partial [D_1(n)P(n,t)]}{\partial n} + \frac{\partial^2 [D_2(n)P(n,t)]}{\partial n^2}, \quad (3.1)$$

where $P(n, t)$ is a density of the distribution function, and the coefficients of drift and diffusion are defined by functions (2.6)

$$D_1 = \frac{1}{\gamma} \left\{ f + \left[M_0(t) \frac{g^2}{\gamma^2} \frac{\partial \gamma}{\partial n} + M_1(t) g \frac{\partial g}{\partial n} \right] \right\}, \quad (3.2)$$

$$D_2 = M_0(t) \frac{g^2}{\gamma^2},$$

At the same time the moments of the correlation function (2.4) are defined as follows

$$M_0(t) = M_0 = I,$$

$$M_1(t) = M_1 = I\tau.$$

In the stationary case $P(n, t)$ does not depend on the time, and then the steady-state of the corresponding process (condensation, disassembly or evaporation) is defined by the extremum of the distribution, which reduces to the expression

$$J_{ac} = (n - n_e)(1 + n^2) - I\tau\sigma(1 + \kappa)^2 n + \frac{2I\sigma(1 + \sigma)(1 + \kappa)^2 n}{(1 + \sigma(1 + n^2))^2}. \quad (3.3)$$

The condition $J_{ac} = -n_e$ limits the domain of existence of the zero solution (evaporation process) and follows from equation (3.3) after substituting $n = 0$.

After the numerical analysis of the dependence (3.3), we construct a phase diagram of the system (Fig. 1), which defines the region of the existence of different solutions of equation (3.3).

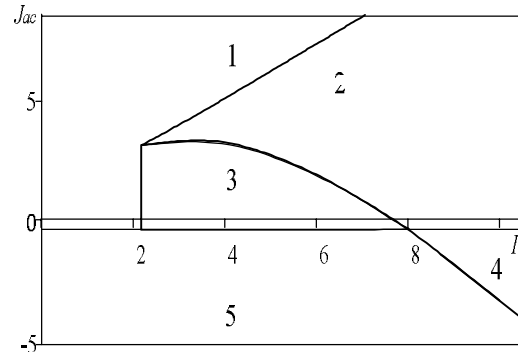


Fig. 1 - Phase diagram of the system for $n_e = 0.25$, $\tau = 0.25$, $J_{ac} = 4$, $k = 0.25$.

As it is evident from Fig. 1 the phase diagram has five regions. The region 1 corresponds to the process of condensation; with a decreasing accumulated flow or with increasing of noise intensity the system falls in the region 2, where the condensation process coexists with the process of the surface disassembly, and equation (24) has two nonzero solutions. In region 3 only the state of disassembly is realized. Here the condensate atoms, previously deposited on the substrate, do not completely evaporate, forming a nanoporous structures. For the increasing fluctuation intensity and reversal accumulated flow the system may move into the region 4, where the evaporation is more intense, coexisting with the island porous structure. In region 5 only the evaporation process is realized.

4. CONCLUSION

The study of the stochastic plasma-condensate system was performed based on the synergetic scheme that takes into account the self-consistent behavior of

the concentration of adsorbed atoms, the temperature of the growing surface and desorption flow. At the same time to describe the conditions maximally close to the experiment the temperature and flow fluctuations were taken into account. Solving the corresponding Fokker-Planck equation in the stationary case, the phase diagram of the system was constructed. The appropriate

regions of the diagram corresponding to the condensation, disassembly and evaporation processes were analyzed. As a result, we can conclude that the represented investigation is quite relevant for the consideration of the conditions for the obtaining of the nanoporous materials.

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