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SELF-ORGANIZED WORLD OF PLASMA NANOSCIENCE: APPROACHES TO NUMERICAL SIMULATION OF COMPLEX PROCESSES ON LOW TEMPERATURE PLASMA EXPOSED SURFACES

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Key words: plasma, magnetron, microwave, arc, nanoscience, nanostructure, self-assemby

Abstract: In this paper we consider several general questions and problems related to self-assembly and self-organization on plasma-exposed surfaces. We are dealing here mainly with the case of low temperature plasma that can be produced in magnetrons, vacuum arcs and microwave sources. Such kind of plasma is very promising environment for the advanced nanotechnology, and its potential in promoting and controlling self-assembly and self-organization on plasma exposed surfaces is very large. Recent investigations, referenced in this work, have shown that the self-assembly and self-organization in low-temperature plasmas can be very effective and versatile tool for the production of complex nanostructures on surfaces. We discuss here several problems and possible features of self-organization in low-temperature plasmas, and indicate some possible ways for the future development and investigations.

Spontana organizacija v svetu plazemske nanoznanosti: pristopi k numerični simulaciji procesov na površini materialov, ki so izpostavljeni nizkotemperaturni plazmi

Kjučne besede: plazma, magnetron, mikrovalovi, oblok, nanoznanost, nanostrukture, spontana organizacija

Izvleček: V prispevku obravnavamo nekatera splošna vprašanja, ki se pojavljajo pri razumevanju procesov spontane organizacije atomov na površini materialov, ki so izpostavljeni nizkotemperaturni plazmi. Osredotočimo se na plazmo, ki jo ustvarimo z naslednjimi razelektritvami: magnetronsko, mikrovalovno in obločno. Takšna plazma predstavlja obetavno okolje za sodobne nanotehnologije, saj omogoča širok izbor parametrov, ki so potrebni za vzpodbujanje rasti nanostruktur in spontano organizacijo atomov na površini različnih materialov. V prispevku povzemamo rezultate najnovejših raziskav, ki jasno kažejo, da je s tovrstno plazmo mogoče vzpodbuditi rast dobro orientiranih nanostruktur, ki imajo lahko zelo zapleteno strukturo. V prispevku tudi komentiramo navajamo in komentiramo nekatere težave, s katerimi se soočamo pri sintezi tovrstnih materialov in nakazujemo možne prihodnje poti raziskav na tem področju.

1 Introduction

The methods of synthesizing complex large-scale nanodevices and extra-large arrays of nanostructures /1, 2/, large arrays of carbon nanotubes /3, 4/ and nanowires / 5, 6, 7, 8/, nanoparticles /9/, various other nanostructures /10, 11, 12/, nanofiber /13/, nanotips /14/ and nanobelts /15, 16/ based on self-organization and self-assembly of complex nanosystems on low temperature plasma-exposed surfaces is one of the most promising ways for synthesizing complex surface-based nanosystems. This technology may be also affectively applied to synthesizing various coatings /17, 18/, nanowire-based probes /19, 20/ and sensors /21, 22, 23/, for bacteria degradation and other biology-related issues /24, 25/. The low-temperature plasma and plasma-based processes are very suitable for various techniques of nano-synthesis /26, 27, 28, 29/, for functionalization /30, 31/ and treatment /32, 33/ of various structured materials and systems /34/, including textile /35, 36/, fabrics /37, 38/, nano-membrane systems /39/ and various materials such as polyolefines /40/, polyamides /41/, graphite-polymer composite /42/, nanocomposite diamonds /43/ and nanotubes /44/. The microwave plasma and microwave plasma - based equipment /45, 46/, as well as magnetrons /47, 48/, setups with radio-frequency inductively coupled plasmas /49, 50/ and vacuum arc setups /51/ appeared to be the most promising environment and setups for these processes /52, 53/. Precise control of self-organization is very important for all processes and all techniques, but some applications such as those based on complex-shape nanowires /54/, as well as nanowires for solar cells /55, 56/ require especial attention to the controllability; as a result, researchers pay a great attention to the characteristics, regimes, and properties of the plasma-generating equipment /57, 58/.

In this situation, the studies on general question of self-assembly and self-organization on plasma-exposed surfaces are of greatest interest /59, 60/. The complex sophisticated methods of numerical simulation allowing detailed investigation of the entire process of self-organization are particularly promising and should be extensively developed /61, 62, 63/. The progress in this direction is significant /64, 65/, but a need in designing new complex numerical methods is pressing. Taking this into account, we have developed several numerical approaches and techniques able to simulate self-organizational and self-assemble processes on plasma-exposed surfaces. This approach allows modeling the whole system consisting of plasma bulk and surface.

The paper is organized as follows. In Section 2, we introduce several typical groups of nanostructures that can be produced by self-organization and self-assembly on plasma-exposed surfaces, and point out some major difficulties that appear in modeling self-organization. In Section 3, we describe an approach and complex model that can be used for numerical simulation of the self-organization and self-assembly in nano-world; in Section 4, we consider briefly the main numerical approaches, boundary conditions and some typical results obtained by numerical simulation of the self-organization and self-assembly on plasma-exposed surfaces.

2 Typical self-assembled and self-organized structures on plasma-exposed surfaces

In general, all self-assembled and self-organized nanostructures may be conditionally divided to the five main types: vertical and vertically-aligned nanostructures such as vertical nanotubes, nanorods, nanotips /66, 67, 68/; one-dimensional nanostructures (nanodots, quantum dots) /69, 70/; two-dimensional nanostructures such as nanowalls /71/; complex self-assembled nanosystems and nanomaterials such as nanowires formed between surface-grown nanodots /72, 73/; nanostructures of complex internal structure, for example, nanodots with core-shell structure and partially-saturated nanoparticles /74, 75, 76/. All these types are very important for the present day nanotechnology, and all of them represent significant difficulties in modeling, especially in cases when the self-organization processes are involved. It is important to note that modeling of low-dimension nanostructures (one- and two-dimensional, i.e. nanodots and nanowalls) cannot be considered as easier task, since in any self-organization process the number of objects involved is very large, and thus the complex (actually, 3D) systems should be modeled, even in cases when it consists of 1D nanodots. Besides, the models capable of describing the self-organization on surface should involve sub-models of several levels and hence several different scales; all this makes this problem very complex.

3 Approach to model self-assembled and self-organized nanostructures

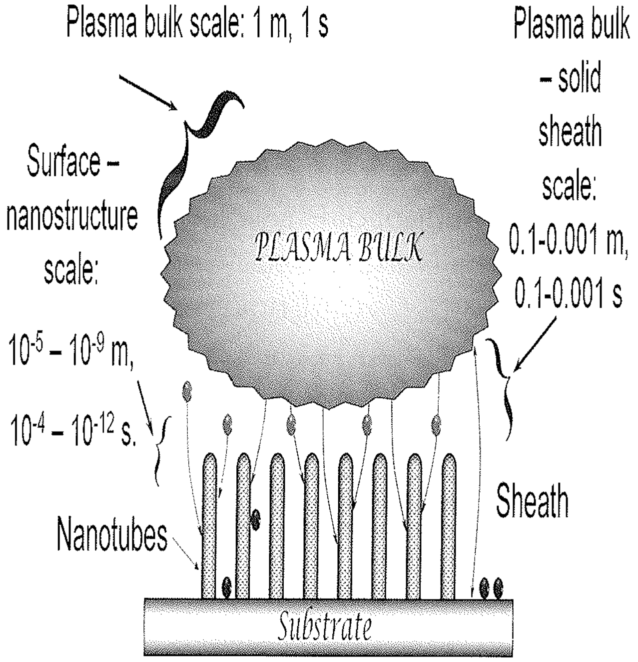

Fig. 1: Three levels of processes in self-assembly on plasma-exposed surface

To build an approach and model for simulation of the self-organization on plasma-exposed surface, one should consider at least three scale levels (Fig. 1). In general, they can be characterized as Plasma bulk level (1), Sheath level (2) and Nanostructure level (3); the characteristic sizes are shown in the figure. Thus, the complex model should involve processes and comprise equations for all three levels.

At the first level, the ion current extraction from the plasma bulk is the main process that determines the ion current density to the nanostructures and substrate surfaces, and hence it determines the kinetics of the self–organization and self-assembly, as well as temperature and other macro-parameters.

When the surface is biased, it is separated from the plasma bulk with a sheath, and the electric field is mainly concentrated in this sheath. In the models, it is often assumed that the ions enter the sheath with Bohm velocity that is calculated as *Vb=(Te/mi)1/2*, where *Te* is the electron temperature, and *mi* is the ion mass. Depending on the parameters, the two main cases can be considered, with respect to the relation between the surface potential *US* and the electron temperature *Te*. When the electron temperature is small, the sheath width is calculated:

$$\lambda_S = \frac{\sqrt{3}}{2}\lambda_D\left(\frac{2U_S}{T_e}\right) \qquad (1)$$

where $\lambda_D = (\varepsilon_0 T_e / ne)^{1/2}$ is the Debye length, ε_0 is the dielectric constant, *n* is the plasma density, and *e* is the electron charge.

In opposite case, i.e. when the surface bias is low, the sheath width can be calculated by the relation for Debye lengths:

$$\lambda_S = k_\lambda \lambda_D = k_\lambda \sqrt{\frac{\varepsilon_0 T_e}{ne}}, \qquad (2)$$

where *kl* is a constant of 1-5.

In the sheath, i.e. at the second scale level, the ions move under effect of the electric field created by the biased (charged) nanostructures and biased substrate surface. This electric field has a complex structure, and thus the trajectories of the ions are complex curves, with the ions being deposited onto side surfaces of the growing substrate. In general approximation, the electric field may be calculated as

$$\mathbf{E}(r) = \sum_1^N \left[\int_{S_i} \frac{\rho_i dS}{4\pi\varepsilon_0 r^3} \mathbf{r} \right] + U_S \left(\frac{z}{\lambda_S} \right)^{\frac{1}{3}}, \qquad (3)$$

where *N* is the number of nanostructures, *z* is the distance to the substrate surface, *ri* is the surface density of electric charges on nanostructure surfaces, and *Si* is the surface area of the *i*th nanostructure. The ion trajectory can be calculated by integration of the equation of ion motion in the electric field. With the density of ion current calculated, the flux of ions and atoms to each specific nanostructure can be obtained, and thus the growth kinetics can be calculated.

At the third scale level, i.e. on the substrate and nanostructure surfaces, the equations based on surface and bulk diffusion should be used. Namely, The two-dimensional flux of adsorbed atoms from the substrate surface to the nanostructure can be calculated from the diffusion equation

$$\frac{\partial \eta}{\partial t} = D_S \left(\frac{\partial^2 \eta}{\partial x^2} + \frac{\partial^2 \eta}{\partial y^2} \right) + \psi_{in} - \psi_{vp}, \qquad (4)$$

for adatom density field *h(x, y, t)* on the substrate surface, where *yin* is the external (from sheath) flux of atoms and ions to the substrate surface, and *yvp* is the flux of atom evaporation from the substrate surface. With the diffusion equation solved (in general, only numerical solution is possible due to very large number of boundary conditions involved), the total flux of adsorbed atoms at the border of an individual nanostructure can be calculated from equation:

$$\varphi_i = -2\pi r_i m_a D / \rho \left(\partial \chi / \partial r \right), \qquad (5)$$

where *ri* is the radius of the *i*th nanostructure, *ma* is the adatom mass, and *r* is the density of carbon material. In this case, the surface diffusion coefficient *D* can be calculated as *D = (l2 ´n)exp(-ed/kT),* where *k* is Boltzmann's constant, *ed* is the surface diffusion activation energy, *l* a is the lattice constant of the substrate, and *T* is the surface temperature.

At the third (nanostructure) scale level, the equations for nanostructure growth and reshaping should be added which have the following general appearance:

$$\frac{\partial V_n}{\partial r_{on}} dr_{on} = J_{S_n} dt, \qquad (6)$$

$$\frac{\partial V_n}{\partial h_n} dh_n = J_{e_n} dt. \qquad (7)$$

Here, *ron, hn*, and *Vn* are the base radius, height and volume of *n*th nanostructure; *Jen* is the total flux of ions and atoms to surface of *n*th nanostructure; *Jsn* is the total surface flux of adatoms to the nanostructure from surface, and, finally, ∂*V*/∂*r* and ∂*V*/∂*l* are the nanostructure growth functions (shape- and size-dependent).

The above described model can be used for numerical simulation of the self-assembly processes on plasma-exposed surfaces. However, the three different scale levels of this system represent a very hard problem, which can be solved by involving different numerical techniques at different levels.

4 Numerical approach, boundary conditions and typical results

4.1 Numerical approach and boundary conditions

We have implemented the following numerical techniques for the different levels: at level 1 (plasma bulk), the traditional methods used for the plasma parameters calculation; at level 2 (sheath - surface), the Monte-Carlo (MC) method was used to trace the ions in the electric field created by charged nanostructures; and at level 3 (nanostructure level), depending on the types of nanostructures to be described, the diffusion or Kinetic Monte Carlo techniques.

The total model uses *N+1* boundary conditions, where *N* is the number of nanostructures on surface. Besides, additional boundary conditions should be set at the boundary of simulation domain which is restricted in the numerical process. An initial condition for the ion motion consists in setting the initial ion velocity at sheath border, which can be assumed as Bohm velocity. The initial shapes of the nanostructures can be set as representative for each specific type.

4.2 Typical results

Now, we consider several typical results obtained by the above described model. We recall that our main aim is to demonstrate that the complex multi-scale numerical approach is capable of simulating self-assembly and self-organization processes on plasma-exposed surfaces.

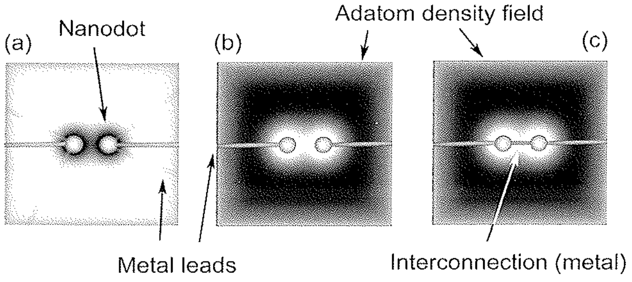

Fig. 2: Formation of metal leads to nanodots and interconnection between nanodots on surface: (a) - formation of external leads to the nanodots; (b) - formation of final leads; (c): formation of metal interconnection between nanodots. The correct growth of leads and interconnections is due to proper selection of the growth conditions.

In Fig. 2 we show the simulated process of the self-organized formation of metal leads (long nanowires shown in green in the figure) to nanodots on surface, and then - formation of metal interconnection (red nanowire) between the nanodots; thus, with the material correctly chosen, this self-assembled system may represent a base for semiconductor nano-diode and nano-transistor.

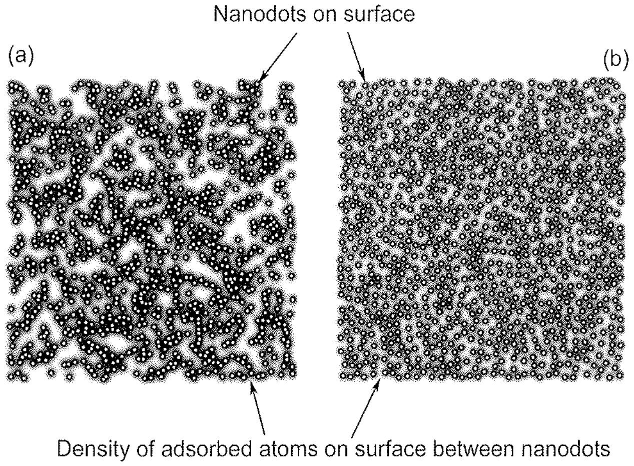

Fig. 3: Self-organization in large arrays of nanodots: formation of ordered pattern: (a) - initial pattern of very low ordering; (b) - final pattern of higher ordering, the nanodots were re-arranged due to self-organizational processes on surface.

In the figure, the first fragment (a) illustrates the formation of leads; the dimmed green leads are not preferable by the conditions of growth, and eventually the process results in the perfect oppositely directed leads (b). Then, with the density of adsorbed atoms on surface increased (as seen from darker color), the interconnections of other metal can be formed directly between the nanodots. The correct growth of leads and interconnections is due to proper selection of the growth conditions and correct guidance of the self-organization.

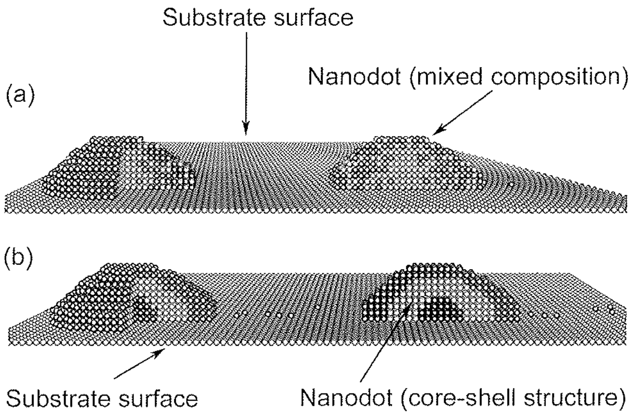

Fig. 4: Self-assembled core-shell nanodots on plasma-exposed surface: (a) - Nanodot of mixed composition, i.e. with the external layer enriched with the dopant, but perfect core-shell structure was not formed; (b) - perfect core-shell structure was formed due to correctly guided self-assembly processes.

The experimental investigation of the interconnection formation on plasma-exposed surface is described elsewhere /lxxiii/.

In Fig. 3 we show the simulated process of the self-organized formation of large array of nanodot on plasma-exposed surface. At the first stage (a), the array of nanodots is completely disordered; at the final stage (b), the pattern is much more ordered. The self-organization processes triggered by the presence of electric field on surface (the nanodots were formed from plasma on biased surface) caused rearrangement of the nanodots and finally resulted in better pattern. The experimental investigation of the ordered nanodot pattern formation on plasma-exposed surface is described elsewhere /lxix/.

In Fig. 4 we show the simulated process of self-assembling the core-shell nanodots on plasma-exposed surface. In the first case (a), the nanodot of mixed composition, i.e. with the external layer enriched with the dopant, was formed; but perfect core-shell structure was not formed; in the second case (b), the perfect core-shell structure was formed on the nanodot due to correctly guided self-assembly processes.

In Fig. 5 we show the results of numerical simulation of self-assembling process that leads to the formation of complex nano-structure consisting of three metal nanodots, interconnected with metal nanowires.

This structure, which can be a prototype of the nano-circuit, was successfully simulated by the above described model.

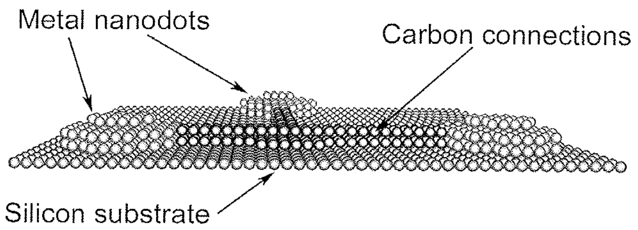

Fig. 5: numerical simulation of self-assembling complex nano-structure consisting of three metal nanodots, interconnected with metal nanowires.

5 Conclusions

In this paper we have discussed some important questions related to the self-organization and self-assembly of nanostructures on plasma-exposed surfaces. We have demonstrated that the proper selection of the structure of model, as well as numerical methods ensures effective modeling and simulation of the complex process of self-assembly and self-organization on low temperature plasma exposed surfaces.

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