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Self-assembly of condensates with advanced surface by means of the competing field selectivity and Gibbs–Thomson effect

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ABSTRACT

Copper and silicon layers were deposited using the accumulative plasma-condensate system. Their surface was found to possess the complex developed morphology using SEM technique. Competing processes of the field selectivity and Gibbs–Thomson effect are considered to describe the formation of the surface. The mathematical model is created on the basis of these effects which describes self-assembly of the surface at the form of adjoining elements of an elliptic section. The comparative analyses of theoretical and experimental results are given.

1. Introduction

Currently, self-assembly of low-dimensional structures by means of deposition under conditions of proximity to thermodynamical equilibrium is of high research interest [1–3]. Until now, only few studies of the structure formation principles have been focused namely on the quasi-equilibrium conditions and their consequences. Therefore, there are still uninvestigated mechanisms of self-organization of morphologically complex growing surface which are based not only on near-equilibrium condensation requirements, but also on focusing of ion fluxes onto projecting parts of the growth surface. We consider using the term "field selectivity" to determine the lastly named factor in further. Therefore, the field selectivity means the idea that ionized deposited atoms being are selectively fixed on projecting parts of the growth surface at the presence of the electric field above it. Our experiments have shown that structures obtained under field selectivity conditions can correspond to coupled columnar elements (an example will be given further in the Section 2 of the present study). Thin film columnar structures known from literature [4,5] do not conform to ours either by morphological characteristics or growth conditions.

So, in the present stage, we set the problem to determine the central mechanisms of structure formation under the field selectivity. The next section of the article covers the peculiarities of the experiment with an explanation of the physical principles of film growth. The third section is devoted to the creation of the mathematical model which describes self-assembly of the developed surface of condensates under the field selectivity. Possible variants of self-assembled surface morphology are discussed. The final section briefly concludes the study.

It should be noted that the obtained structures are of certainly applied interest due to their properties which, however, do not lie at a focus of the present work. Some characteristics of the structures such as electroconductivity are determined by contact phenomena between the columnar elements to a large extent. Therefore, the technological approach offered in the present work can be useful for producing sensors operating in various chemically active medium. Besides, the preliminary tests have shown that the copper condensates change their electrical resistance approximately to 10% during bending the substrate that is due to contact loss between columnar elements and can be used in tensometry.

2. Experimental device and microstructures obtained

In order to perform corresponding experiments, the accumulative plasma-condensate system (APCS) was used [6]. Such devices consist of the magnetron sputterer and the hollow cathode 5 placed in the middle of the magnetron (see Fig. 1).

The material was condensed on the glass substrate 6 situated in the hollow cathode 5 on the water-cooled holder 7. The operational principles of the APCS are based on self-organization of

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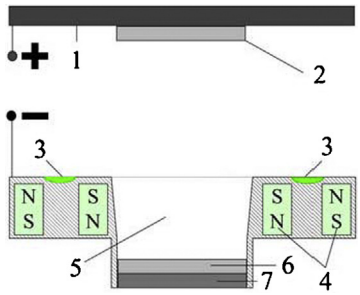

Fig. 1. Cross-section of the axisymmetrical accumulative plasma-condensate system (APCS) (1 – anode; 2 – substrate; 3 – erosion zone; 4 – magnet system; 5 – hollow cathode volume; 6 – substrate; 7 – chiller).

condensation conditions near thermodynamical equilibrium. The mathematical model of self-organization of critically low steady-state supersaturation and physics of the operation principles of the APCS are discussed in detail in Ref. [6]. Due to the fact that the deposited substance localized inside the hollow cathode 5 is considerably ionized [7], and also the negative potential is supplied to the growth surface, there are prerequisites for field selectivity.

The necessity to create the mathematical model describing self-assembly of the developed surface of condensates comes from our corresponding experiments concerning deposition of Cu and Si using the APCS. Argon was used as the working gas during the material deposition. Argon was undergoing deep cleaning directly inside the vacuum chamber during all the technological process by the procedure given in Ref. [8]. We used non-running gas inflow, i.e. the vacuum chamber was cut off from the pumping system after pump-down. After that the chamber was filled with argon to its working pressure P_{Ar} = 6–15 Pa. Then two additional magnetron sputterers of titanium shielded from the APCS were turned on. During titanium sputtering residual chemically active gases were absorbed. Thus, their partial pressure was minimized to the level of about 10^{-7}–8×10^{-8} Pa [8]. The discharge power input P_w was 6–21.9 W and the ratio of the cathode inlet hole area to the cathode inner surface area (see [6]) was 0.5.

The examples of self-assembled developed surfaces of Cu and Si at the form of adjoining convex and concave ellipsoids of revolution are shown in Fig. 2. XRD investigations of the condensates revealed well defined texture of nucleation and growth for Cu ((1 1 1) being parallel to the substrate surface). Meanwhile, the Si condensates possessed amorphous structure. We have assumed this to be caused by primary origination of Si condensate in amorphous form on structurally isotropic glass substrate. The atom-by-atom attaching to the base amorphous layer occurs near thermodynamical equilibrium. That growth process complicates transition to the crystalline state. Vicinity to the phase equilibrium promotes the maximal filling of chemical bonds that are the prerequisite for quasi-crystalline structure formation.

As it can be seen from the given examples of the developed surface (see Fig. 2a and c), only individual projections and hollows exist on the growth surface during the first stage of self-assembly. However, sufficiently long condensation leads to statistically homogeneous morphology (see Fig. 2b and d) that indicates on the features of self-organization. It should be also mentioned that the field selectivity is possible if the growth surface possesses initial local curvature. Such deviations from perfectly flat surface exist on a substrate and can be also formed during condensate growth.

3. Mathematical model of self-organization of the advanced condensate surface

3.1. Physical basics of the surface growth process

Our aim is not to study kinetics of atomic processes but to follow the transformations of structural elements on macro level. We suppose that adatoms diffusion processes are not crucial in

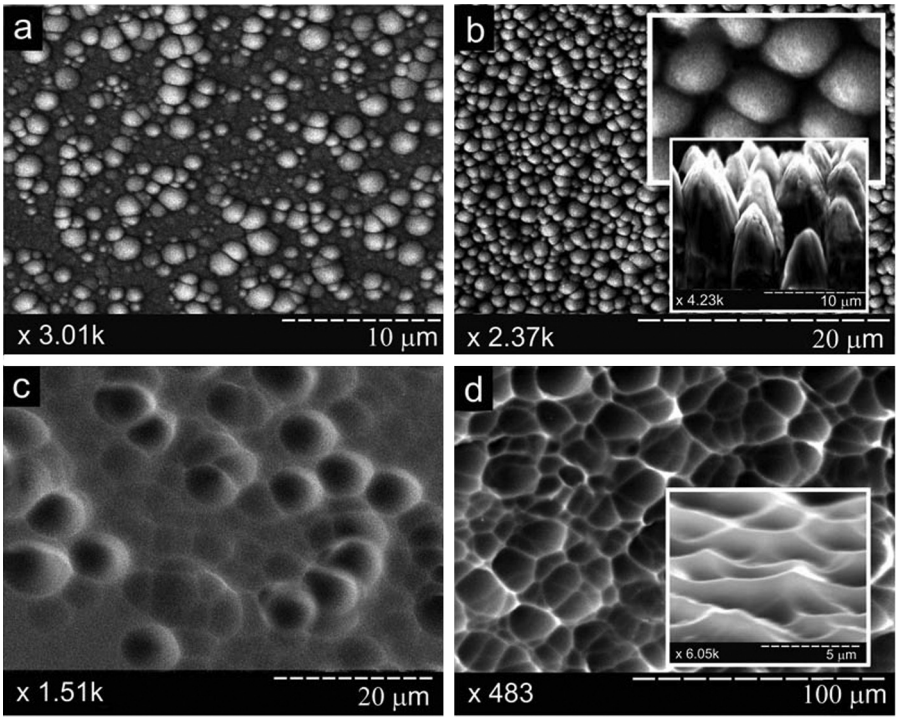

Fig. 2. SEM images of surface morphology of Cu and Si condensates obtained *ex-situ* on glass substrates. (a) Cu, deposition time is 30 min, P_{Ar} = 15 Pa, P_w = 16.8 W; (b) Cu, deposition time is 6 h, P_{Ar} = 15 Pa, P_w = 16.8 W; (c) Si, deposition time is 2 h, P_{Ar} = 6 Pa, P_w = 3.4–6 W; (d) Si, deposition time is 8 h, P_{Ar} = 6 Pa, P_w = 3.4–6 W. Small insertions represent enlarged or lateral view of the samples.