Roughness of 3C-SiC thin film coating computer simulation

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Abstract. This paper deals with efficiency of weakly ionized gas-wall interaction which leads to follows: melting surface, clustering of vapor and melted charged drops nucleation to the solid phase. It shapes roughness of the surface coating.

Keywords: 3C-SiC, thin films, kinetic theory, stochastic equations.
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INTRODUCTION

The charging of the spherical SiC melted drops is possible into weakly ionized gas of low Earth orbits due to high velocity of gaseous particles. The 3C polycrystalline of SiC coating redeposition on Si(100) substrate is studied using stochastic computer simulation /SS/ of phase transition /PT/[1]. SCS allows to model nucleation of SiC spherical melted clusters- drops sizes evolution, its precipitation on Si(100) substrate and formation of SiC cubic shaped crystals nuclei. Kinetic model is based on both: stochastic nature of particles clustering and clusters Brownian motion which is produced by the long-range potential of clusters interaction. Mathematical model of nucleation is described sufficiently by Kolmogorov equation. The SCS advantages are both: Kinetic quasi-linear partial differential equations for nucleation solutions and nuclei Brownian motion model. Quasilinear kinetic equations (Fokker-Planck-Komogorov, Einstein-Smolukhovskii) solution using new (stable and effective) computer simulation method, which is based on the strict results of probability analysis of mathematical physics equation, i.e. using the solution of Ito-Stratonovich stochastic differential equations. Stochastic analog equations ensure [1-3] are of high accuracy. New results of kinetic theory concerning of nuclei kinetic distribution functions /DF/ versus its sizes are calculated. Model can to account: melting of the SiC, drops evaporation, vapor redeposition as well as gases heterogeneous recombination and surface erosion. The rates of this processes depend on asurface roughness formation. Self-organization aspects of nucleation into the “open” plasma-like media taking into account the role of the both: sizes dependent Gibbs free energy of cluster formation and alternating-sign long-range potentials of cluste indirect elastic interaction [4,5]. The deposition stochastic model of SiC nano-islands in forms of charged drops or of crystals is presented as a heterogeneous first-order PT at fluctuation stage [1]. The fluctuation instability of several PTs of "vapor-melt-crystal" plays a crucial role in evolution of DFs versus its sizes. Silicon carbide (3C-SiC) cubic shaped crystals sizes depends on charge value of melted droplets as well as on plasma parameters into discharge.

STOCHASTIC MODELS AND EQUATIONS

Stochastic simulation /SS/ of clustering is performed within molecular kinetic theory and SS is different from imitation modeling (molecular dynamics /MD/) in that the SS calculation of the trajectories of random processes reflects the estimation method for the statistical properties of a particle ensemble in the phase space rather than the Newtonian dynamics of real atoms and molecules which are used in MD. From the standpoint of numerical modeling, the formation of clusters/defects can be represented by the stochastic dynamics of a system with a large number of degrees of freedom. SS method can by used in a state different from the equilibrium state, the space–time scales of which correspond to the evolution level of kinetic DFs in multidimensional phase spaces. The macroscopic characteristics of the medium can be obtained as nonequilibrium DF moments, which distinguish SS from MD and

Monte Carlo calculations. The kinetic approach can be useful if it is important to reveal the causes of instability in the system. Kinetic equations for fluctuation nucleus formation (Kolmogorov–Feller kinetic equation) and for Brownian motion (Einstein– Smolukhovsky equation) are integrodifferential partial differential equations and its were published in previous papers of authors [1-3].

Let us present integral representation of the Ito stochastic differential equation in Stratonovich form for only one stochastic dynamic variable (cluster size g) in point on solid surface $\boldsymbol{r}$={x,y}:

$$
\begin{aligned}
& X(t) = X(t_0) + \int_{t_0}^{t} H(\tau, X(\tau))d\tau + \int_{t_0}^{t} \sigma(\tau, X(\tau))dW(\tau). \\
& H_g = -\frac{1}{kT} D_g(g,t) \frac{\partial \Delta\Phi(g,x,y,t)}{\partial g} - \frac{1}{2} \frac{\partial D_g(g,t)}{\partial g}, \\
& \sigma_g = \frac{1}{q} \sqrt{2D_g(g,t)}, \\
& D_g = D_{g0} g^{2/3} \\
& t_0 \le t \le T_{finish}, \quad g(t_0) = g_0 \in [g_{\min}, g_{\max}], \; g(t) > 2,
\end{aligned}
\tag{1}
$$

where $\Delta\Phi(g,\boldsymbol{r},t)$ is the nucleus formation thermodynamic potential (or the Gibbs energy). T is temperature of vapor in plasma discharge for simulation of charged spherical SiC melted drops and it is substrate temperature for simulation of non-charged SiC germs on surface. Vapor temperature is more than substrate temperature on 300 K. In eq. (1) T_{finish} is the fluctuation stage duration; $g_{\min}$ and $g_{\max}$ are the minimum and maximum values of the initial cluster size (g_0), which are chosen from the region of fluctuation-unstable values of g such that $\Delta\Phi(g_{\min}) = \Delta\Phi(g_{\max}) = \Delta\Phi(g_{cr}) - kT$, where g_{cr} is the critical nucleus size defined in thermodynamics by the expression (2). Here dW is increment of the Wiener random process, q is intensity of random process. The diffusivity $D_g = D_{g0}g^{2/3}$ is characteristic of nucleus formation process of clustering in the phase space of islands (clusters) sizes, where g∈{G}, D_g is functional coefficient (1). For each mentioned model of PT the Gibbs energy has form:

$$
\Delta\Phi = \begin{cases}
\Delta\Phi_{\text{vol}} + \Delta\Phi_{\text{surf}} + \Delta\Phi_{\text{z}} + \Delta\Phi_{\text{ch}} = \Delta\Phi_{drop} & \text{melted spherical drops in plasma discharge} \\
\Delta\Phi_{\text{vol}} + \Delta\Phi_{\text{surf}} + \Delta\Phi_{r} = \Delta\Phi_{am} & \text{amorphous clusters on solid surface} \\
\Delta\Phi_{\text{vol}} + \Delta\Phi_{\text{surf}} + \Delta\Phi_{r} + \Delta\Phi_{\text{edge}} + \Delta\Phi_{\text{edge el.}} = \Delta\Phi_{cr} & \text{crystal nucleus}
\end{cases}
\tag{2}
$$

where $\Delta\Phi_{\text{vol}} = -a_{\Phi}g$ is connected to the difference between chemical potential of vapor and melt phase, vapor and crystalline and other, $\Delta\Phi_{\text{surf}} = bg^{2/3}$ is connected to the change in the surface free energy of the system, $\Delta\Phi_z = cg^{-1/3}$ corresponding with charge on SiC melted drops in plasma (or ionized gas). The chemical reaction on germ surface in presented model is considered as additive part in Gibbs energy accounted as $\Delta\Phi_{\text{ch}} = -a_{chem}g^{2/3}$

The differences between the energies of nucleus formation over the site or interstice of the substrate lattice are accounted by

$$
\Delta\Phi_r = \frac{\Psi}{\sqrt{g}} \left(2 - \cos\left(\frac{2\pi x}{a_x}\right) - \cos\left(\frac{2\pi y}{a_y}\right)\right) \tag{3}
$$

The short-range contribution of edges to the Gibbs energy $\Delta\Phi_{\text{edge}} = d_1 g^{1/3}$ also has an effect on the crystal cluster formation. The contribution of cluster edges to the elastic relaxation energy is given by $\Delta\Phi_{\text{edge}} = -d_2 g^{1/3} \ln(g)$.

If at precipitation time $\Delta\Phi_{\text{drop}} \ge \Delta\Phi_{\text{cr}}$ that crystal 3C-SiC nuclei format, if $\Delta\Phi_{\text{drop}} \ge \Delta\Phi_{\text{cr}}$ that amorphous clusters format. Amorphous germs have globe calotte forms with wetting angle according to Yung law.

The potential of the interaction of nuclei with each other and lattice defects is long-range and sign-alternating; the interaction is realized via the perturbation of lattice acoustic phonons and Friedel oscillations of the electron density (for metals and semiconductors). The potential of indirect long-range interaction is similar to that used in [4,5]. To solve the system of interconnected SDEs (SDE for cluster size g and SDE for $\boldsymbol{r}$ space coordinates) with functional coefficients (modified [1-3] for quasi-linear case Artem'ev method [6] of SDE's solution) is used. This method is no

lower than of the second order of accuracy in the root-mean-square deviation. In difference from the Euler scheme of algorithms widely used in MD presented here the Artem'ev method is stable and effective due to the using of Stratonovich form of to SDE. For the *i*th trajectory of the diffusion MP, the values of g_{n+1} at the instant $n+1$ are calculated using the formulas

$$g^i_{n+1} = g^i_n + \left[I - \frac{h_g}{2}\frac{\partial H^i_n}{\partial g^i_n} \right]^{-1} \left[h_g H^i_n + \sqrt{h_g}\sigma^i_{ng}\xi^i_{ng} + \frac{h_g}{2}\frac{\partial \sigma^i_{ng}}{\partial g^i_n}\sigma^i_{ng}(\xi^i_{ng})^2 \right],$$

$$H^i_{ng} = -\frac{1}{kT} D^i_g\left(g^i_n, t_n\right)\frac{\partial \Delta\Phi^i_n\left(g^i_n, x^i_n, y^i_n, t_n\right)}{\partial g^i_n} - \frac{1}{2}\frac{\partial D^i_n\left(g^i_n, t_n\right)}{\partial g^i_n},$$

$$\sigma^i_{ng} = \frac{1}{q}\sqrt{2D^i_n\left(g^i_n, t_n\right)}.$$

Here, I is the unit matrix, h_g is the time step for the size evolution, ξ is random function modeling Winer process, g_n^i is the solution to SDE at the grid point, which corresponds to the time t_n and trajectories i.

RESULTS

On fig. 1 is presented kinetic DF of clusters versus its sizes which was calculated for case of refrigerated Si(100) surface. DF from rib cub size can be considered as characteristic of roughness of surface. Vapor temperature T =2100 K in plasma discharge and cluster density $3 \cdot 10^5 cm^{-3}$. The dimensionless time has value 10^{-4} s. Upstream plasma density is equal to 10^{14} cm^{-3}. Substrate temperature is 1600 K. Three maximums are observed on DF. First maximum connects with big number of small clusters, second maximum corresponds to increased 3C-SiC nucleus. These two maximums exist for all possible temperatures. Third maximums appear if vapor temperature in plasma discharge more 1900 K.

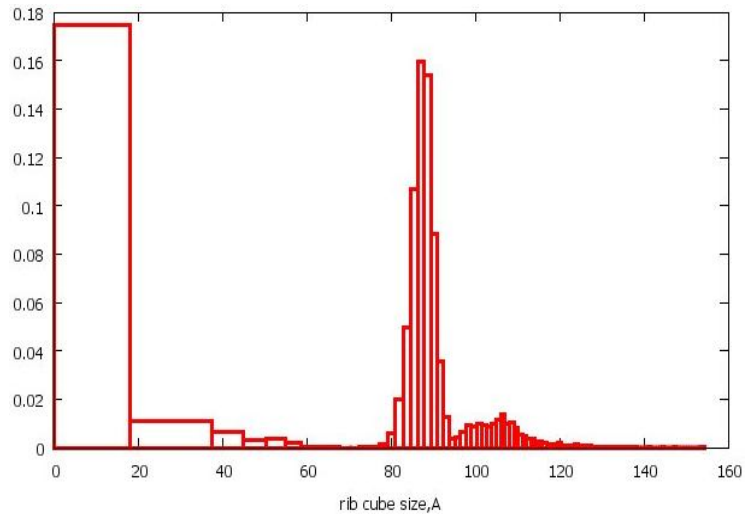

Figure 1. The dependence of clusters DF of non-charged 3C-SiC on Si(100) from rib cube size in A.

The average rib size can be considered as characteristic of roughness of surface. The dependence of average rib size from charge density on SiC melted drops (x-age) and vapor temperature in plasma discharge (y-axe) is presented on figure 2. The charge density on SiC melted drops is assumed constant during each computer experiment. In examined case charge on SiC drop ~ $g^{1/3}$. Si(100) substrate temperature (and SiC cluster temperature after precipitation on Si(100) surface) is less then vapor temperature in plasma discharge on 300K.

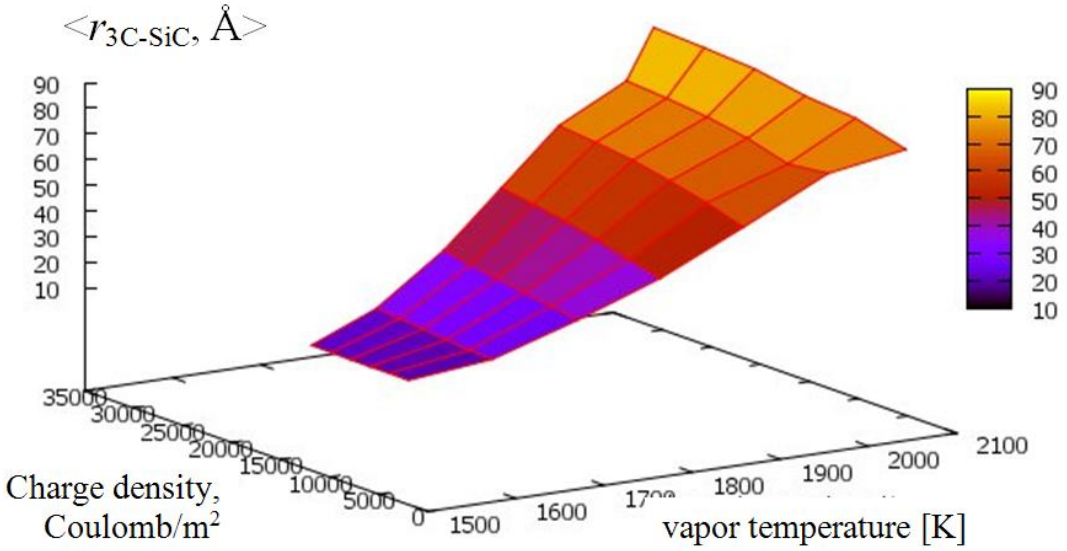

Figure 2. The dependence of average rib size from charge density on SiC melted drops (x-age) and vapor temperature in plasma discharge (y-axe) is presented on this figure.

CONCLUSION

At the high temperature (beginning 2000K) and high charge density distribution function /DF/ of 3C-SiC on Si(100) has 3 character rib sizes. For temperature 2100K these are 2 nm, 9nm and 11 nm after 10^{-4} s since precipitation on Si(100). If charge density decreases that third maximum (corresponding big rib sizes) disappears. At the low temperature (~1900K) DF of nucleus clusters has only two maximums. For temperature 1800K character rib sizes are 2 nm and 5 nm.

Powder of 3C-SiC DFs are non stationary, which depend on follows: charging of melted droplets, its stoichiometric state, neutralization and coagulation on the substrate, model jump of temperature etc.

Silicon carbide (3C-SiC) cubic shaped crystals sizes depends on charge value of melted droplets as well as on plasma parameters into discharge.

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