Keldysh Institute • Publication search

Keldysh Institute preprints • Preprint No. 149, 2019

ISSN 2071-2898 (Print)
ISSN 2071-2901 (Online)

Zmievskaya G.I.

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***Recommended form of bibliographic references:*** Zmievskaya G.I. Structures of Self-Organization of Porosity in Solid // Keldysh Institute Preprints. 2019. No. 149. 15 p. doi:10.20948/prepr-2019-149-e
URL: http://library.keldysh.ru/preprint.asp?id=2019-149&lg=e

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**ИНСТИТУТ ПРИКЛАДНОЙ МАТЕМАТИКИ**
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**G.I. Zmievskaya**

# Structures of Self-Organization of Porosity in Solid

**Москва — 2019**

**Змиевская Г.И.**

## Структуры самоорганизации пористости в твердом теле

Компьютерное моделирование зарождения пористости при имплантации ионов инертного газа проведено на основе уравнений кинетической теории начальной стадии фазового перехода — флуктуационной нуклеации. Стохастическая молекулярная динамика, использующая систему уравнений Ито в смысле Стратоновича, позволяет анализировать образование кластеров вакансионно-газовых пор в зависимости от их неравновесных функций распределения как по размерам, так и по декартовым координатам объема слоев. Самоорганизация в фазовом пространстве стохастических динамических переменных возникает при броуновской диффузии вследствие дальнодействующего коллективного упругого взаимодействия пор в решетке при условии «открытой» физической системы имплантации ионов в решетку за короткое время нуклеации (менее 100 микросекунд). Примеры структур представлены для условий флуктуационной неустойчивости фазового перехода в тонком слое карбида кремния при облучении $Xe^{++}$ с энергией ионов 5 -10 кэВ.

***Ключевые слова:*** кинетическая теория, стохастическое моделирование, пористость, карбид кремния, кластеры пор, структуры самоорганизации.

**Galina Ivanovna Zmievskaya**

## Structures of Self-Organization of Porosity in Solid

Computer simulations of porosity nucleation under implantation inert gas ions carried out on the basis of equations kinetic theory of initial stage of phase transition - fluctuation nucleation. Stochastic molecular dynamics, which use set of Ito equation in Stratonovich sense, allows to analyze the formation of structures vacancy - gas pore clusters in dependence on their non-equilibrium distribution functions from both: size and Cartesian coordinates of the volume of the layers. The self-organization in phase space stochastic dynamical variables appears during Brownian diffusion due to long - range collective elastic interaction of pores in lattice under condition "open" physical system of ions implantation in the lattice at short time of nucleation (less to 100 microsecond). The examples of structures are presented for conditions of fluctuation instability of the phase transition in a thin layer of silicon carbide upon irradiation with $Xe^{++}$ with an ion energy of 5 -10*keV*.

***Key words:*** kinetic theory, stochastic simulation, porosity, silicon carbide, pore clusters, self-organization structures.

Работа выполнена при частичной поддержке Российского фонда фундаментальных исследований, грант 18-01-00436.

## CONTENTS



## INTRODUCTION

Simulation of phase transition is considered in a numerical experiment based on the theory of stochastic dynamic variables [1]. Among the processes that modify the functional properties of the surfaces, an important role belongs to the phase transitions of the 1st kind [2] such as condensation of vapor in the volume of discharge plasma on the surface of the substrate [3], formation of defects consisting of vacancies and gas (vacancy gas) pores /VGD/ in the crystal lattice of the material [4,5], crystallization, melting, and many other phenomena of the interaction of the plasma with the surfaces [6], devices, installations, etc. At the initial stage of phase transitions with characteristic times of ~ 100 μsec, nuclei of a new phase have the form of melt droplets during vapor condensation or VGD pores when the gas implants in the crystal lattice.

Implantation of ions into solid is accompanied by the phase transition of the 1-st kind which manifests itself as VGD pores in lattice. Studying mechanisms of nucleation formation of non-point radiation induced defects (or "blisters") implies using the non-stationary kinetic equation (Kolmogorov-Feller and Einstein-Smolukhowski) with non-linear coefficients describing nucleation processes, clustering of nuclei and their Brownian motion in phase space of cluster sizes and Cartesian coordinates in lattice. Stable method of solving Ito stochastic differential equations /SDEs/ in sense of Stratonovich which is used in the analysis of nucleation dynamics was developed earlier. The stochastic dynamic variables of the problem are analyzed as the probability density of random processes or as the distribution function of kinetic equations in partial derivatives of molecular-kinetic theory. The

long-range interaction of vacancy-gas pore nuclei in a unit volume arises due to indirect elastic forces in the lattice, of a collective nature which leads to the creation of pore cluster structures (“pre-cracks” in the samples). Computer simulation is able to be used for non - point defects prediction in lattice.

Molecular dynamics / MD / is a numerical method for simulating on the atomic scale of the movement of implanted particles. Modern ideas about the mechanisms of damage to the crystal lattice by cascades of implanted atoms and the formation of point defects is based on Monte-Carlo simulation. As a result of integration Newton's classical equations analyze the dynamic development of the state system of atoms. A numerical method of stochastic molecular dynamics [3 - 6] of the Monte Carlo family of methods [7] was developed to solve the problems of kinetic theory (in particular a model of non - equilibrium phase transition stage) which is different from MD since it uses stochastic differential equations /SDEs/ as characteristics of motion and the derivation of which used the exact relation of stochastic problem coefficients (a drift and a stochastic diffusion in SDEs) with the coefficients of partial differential equations of kinetic theory following from considered problems and models their coefficients approximation. The choice of algorithms for the SDEs solution is also important. The study of the properties of silicon carbide is relevant [8] in the construction of automotive, aerospace and rocket technology, both in terms of obtaining nano scale powders during vapor condensation for composites, and in the study of radiation damage to protective coatings, where appearance structures of pore leads to micro crakes development.

## NUMERICAL MODELING OF THE NUCLEATION AND POROSITY IN THE SAMPLE

The mechanism of porosity formation is considered as a result of a phase transition at its initial nonequilibrium stage and is associated with the generation of vacancies upon irradiation with inert gas ions, i.e., defects arise in the crystal lattice. The process model is represented by a superposition of two random processes: (1) diffusion in the phase space {G} of the nuclei sizes [2] or clustering of defects, and (2) Brownian diffusion of their centers of mass in a sample consisting of two layers (silicon carbide and metal).T The computational domain is shown in Fig.1. Initially the pores in the layers are uniformly distributed and several initial distributions over sizes are considered (delta - function, equilibrium distribution, et al.)

Defects in the crystal lattice are described by a nonequilibrium size distribution function, their motion occurs under the action of a sum of potentials that take into

account the interaction of defects with each other, with the boundaries of the layers and a number of other interactions, the dependence of the potential on the distance between the defects and the elastic properties of the lattice is derived using the Feynman diagrams taking into account the properties acoustic phonons of the lattice and Friedel oscillations of electron density. Each pore experiences the influence of all neighbors through a change in the vibrations of acoustic phonons when defects appear, which determines the collective nature of the processes in a solid. This means the collective nature of the potential of the interaction between pores. Thus, the potential is recalculated when position of VGDs is changes. Distance between pores, which is needed for their merging or stopping of their motion, determines spatial structures of defects. Conditions of the "open physical system", used in the model, are determined via the thermodynamic parameters of the lattice and implanted particles: ion energy (~5-10*keV*), dose of ion flux, and surface temperature of the sample, each calculation of porosity formation are kept constant during porosity formation in a crystalline lattice [4,5]. Nucleation process is described by the Fokker - Planck - Kolmogorov kinetic equations and by their stochastic analogue – the Ito - Stratonovich equations. Evaluated are the degree of dispersion of inclusions of gas clusters and their structures in layers of thin films, when expose to fluxes of inert gases ions, which are neutralized at the sample border and form the nuclei while interacting with the flow of vacancies.

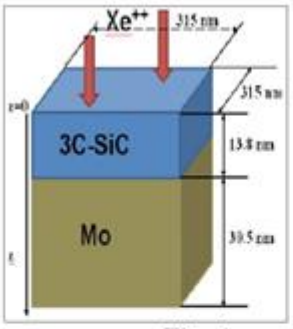



Fig. 1 .

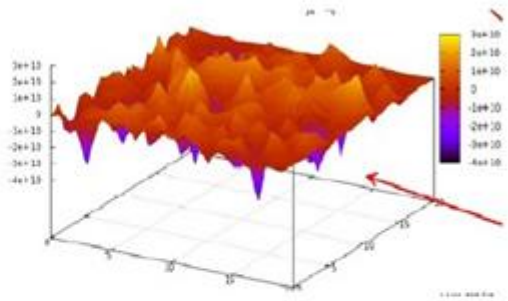


Fig. 2.

Fig.1. Sizes of the sample (*SiC/Mo*) and flux $Xe^{++}$ direction relative to thin film layers (at the left).
Fig.2. Equipotential surfaces of $U(x,y,z)$, (arb. units), of the indirect elastic interaction of pores in the lattice of layer *SiC* , $T/T_{melt}$=0,53, $T_{melt}$ is the melting temperature of *SiC*, the dose of $Xe^{++}$ flux is $10^{15}$ cm$^{-2}$, ion energy is 5*keV*.

Self - organization means formation of ordered structures (similar to ordered turbulence flow) from initially stochastically uncontrolled states without external correcting impact.

## STOCHASTIC MOLECULAR DYNAMIC DESCRIPTION

Numerical experiment for the stochastic molecular dynamics method has the following features:

1. SDEs are formulated, corresponding coefficients are related to the coefficients of kinetic equations; those coefficients should be smooth and twice continuously differentiable, which provides the conditions for the existence and uniqueness of the SDEs solution,
2. discrete model of the medium includes stochastic dynamic variables [1] describing clustering of particles (vacancies and gas); that model is determined by the Gibbs energy model of the nuclei formation, corresponding Brownian motion of those nuclei is determined by the interaction potentials of the fixed nucleus with the other nuclei in the lattices of the sample layers and conservation laws,
3. the choice of the Stratonovich form for the Ito SDEs allows to simplify a representation of stochastic integrals in the solution of the SDEs systems, solution of which replaces the solution of the partial differential equation;
4. the trajectories of the random process should be analyzed in terms of the transient probability density of Markov processes for Kolmogorov equations and to find the kinetic functions of the cluster size distribution in the unit volume and the distribution of defects in the sample in depth for the given moments of time.

## KINETIC AND STOSHACTIC EQUATIONS OF MODELING

Let us consider the model of the fluctuation stage of porosity formation. Processes of clustering are represented by a diffusion in the phase space of cluster sizes {G}. The idea to use a random Markov process allows us to formulate the problem in the form of the Fokker–Planck–Kolmogorov (FPK) equation for the probability density of cluster size DF $f(g,t)$ (source of "monomers" :vacancies and implanted gas) is denoted as $S_a$) [3-5]:

$$\frac{\partial f(g,t)}{\partial t} = \frac{1}{kT}\frac{\partial}{\partial g}\left[ D(g,t) f(g,t) \frac{\partial \Delta\Phi(g,t)}{\partial g} \right] + \frac{\partial}{\partial g}\left[ D(g,t) \frac{\partial f(g,t)}{\partial g} \right] + S_\alpha . \qquad (1)$$

Initial conditions are $f(g,0) = f_0(g,0), \quad \frac{df(g,t)}{dg}\Big|_{g=2} = 2, \quad f(g,t)\Big|_{g=2} = 0.$

Here $g$ is the cluster size or the number of incompressible volumes of gas in the pore, $D$ is the diffusion coefficient in the space of cluster sizes {G}, which depends on temperature $T$ and gas pressure, $D \sim g^{2/3}$; $\Delta\Phi$ is the free energy of nuclei formation

(or Gibbs energy), which depends on the difference of chemical potentials of phases, on surface tension of pores, on elastic properties of the layers and on distance from the boundary between the layers.

The value *f(g, t)dg* determines the number of nuclei in the size range (per unit volume of the medium). The range of variation of variables is $g \in [2, G], \quad t \in [0, \infty]$.

The Einstein-Smoluchowski equation for the Brownian motion of the centers of mass of clusters of defects reads as

$$\frac{\partial f_g(\vec{r},t)}{\partial t} = \frac{\partial \left[ D_r(\vec{r},t) \frac{\partial f_g(\vec{r},t)}{\partial r} \right]}{\partial \vec{r}} - \frac{\partial \left[ \frac{\vec{F}(\vec{r},t)}{M_g \gamma} f_g(\vec{r},t) \right]}{\partial \vec{r}} - Q, \qquad (2)$$

boundary conditions (periodic on *x* and *y*), and on the *z* axis are chosen similarly to [5], *Q* is the runoff of clusters.

Distribution *f(g, t)* from (1) allows to find $M_g$ (clusters' masses) for (2). The force projection on the flow direction z is $F_z - \partial U(x,y,z)/\partial z$, where *U(x, y, z)* is a superposition of the potentials of the indirect elastic interaction of VGDs (pores) in the layers with each other and with the boundaries of the computational domain; *U(x, y, z)* reflects long-range self-consistent interactions VGDs due to long-range elastic forces in lattice associated with the perturbation of oscillations of acoustic phonons by VGDs in the lattice; the distance $\boldsymbol{r} = \boldsymbol{r}(x, y, z)$ between defects is measured in units of interatomic distances in the formula for *U(x, y, z)* are the projection distances on the axis *x, y, z* (see below). The sign of the potential depends on the combination of the elastic moduli.

The model (1) for the stationary phase transition at the initial stage was proposed by Ya. B. Zel'dovich [2], whose ideas were developed in [3 - 6]. Stable algorithms proposed in [3] (see also references there) of the SDEs solution became a tool of computational experiments for the kinetics of non-equilibrium fast - flowing processes. We consider here the problem statement, omitting the description of the algorithms.

The Cauchy problem for the stochastic differential equation in the sense of Ito, corresponding to the FPK (1) equation, has the form:

$$X(t) = X(t_0) + \int_{t_0}^{t} H(\tau, X(\tau))d\tau + \int_{t_0}^{t} \sigma(\tau, X(\tau))dW(\tau).$$

Where $H(\tau, X(\tau))$ and $\sigma(\tau, X(\tau))$ are functional - coefficients of drift and diffusion, *W* is the Wiener random processes. Variable *X* can be represented by stochastic dynamic variables: the size of the clusters *g*, or/and Cartesian coordinates *r={x, y, z}*. To write SDEs in the sense of Stratonovich we replace equation (1) by the equation for X(t) or its stochastic analogue using the appropriate form of coefficients:

$$H_g = -\frac{1}{kT} D_g(g,t) \frac{\partial \Delta\Phi(g,x,y,t)}{\partial g} - \frac{1}{2} \frac{\partial D_g(g,t)}{\partial g},$$

$$\sigma_g = \frac{1}{q} \sqrt{2\mathrm{D}_g(g,t)}, \quad D_g = D_{g0} g^{2/3}$$

Ito's SDEs in the sense of Stratonovich has the form,

$$dg(t) = \left( -\frac{D(g,t)}{kt} \frac{\partial \Delta\Phi(g,t)}{\partial g} + \frac{1}{2} \frac{\partial D(g,t)}{\partial g} \right) dt + \sqrt{2D(g,t)} dw(t), \quad t \in [t_0, T_k]; \; g(t_0) = g_0, g(t) \geq 2. \tag{3}$$

Here *dw(t)* is a model increment of a random process corresponding to (1). Similarly, it is possible to present the drift and diffusion in the SDEs to determine the

coordinates ***r*** of the Brownian particles (pores) in the lattice:

$$H_r = -\frac{1}{\gamma M_g} D_r \frac{\partial U(r)}{\partial r} - \frac{1}{2} \frac{\partial D_r}{\partial r}, \quad \sigma = \frac{1}{q} \sqrt{2\mathrm{D}_r}$$

SDEs corresponding to (2) can be presented as:

$$dr(t) = \left( -\frac{F(r,t)}{M_g \gamma} + \frac{1}{2} \frac{\partial D_r(r,t)}{\partial r} \right) dt + \sqrt{2D_r(r,t)} dw(t), \tag{4}$$

where potential $U_{ij}$ has the form

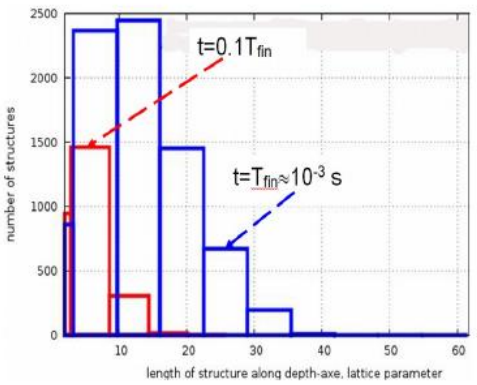
$$U_{ij} = \sum_{i \neq j}^{N} \left( \frac{b_r \left[ 3/5 - \frac{(x_i - x_j)^4 + (y_i - y_j)^4 + (z_i - z_j)^4}{(r_i - r_j)^4} \right]}{(r_i - r_j)^3} + \frac{a_r \cos\left( c_r |\mathbf{r}_i - \mathbf{r}_j| \right)}{(r_i - r_j)^3} \right),$$

Here *i*, *j* are the numbers of interacting pores in corresponding volume being numerate them from 1 to *N*. An example of $U_{ij}$ is presented in Fig.2, coefficients $b_r$, $a_r$, $c_r$ are the model parameters of the respective lattice layers. The correlation between concentrations of vacancy-gaseous defects in layers of sample and the potentials values is established by the corresponding normalization [5]. The distribution of potential in the lattice defines in a consistent way a porosity of the material with taking into account $D_r$, that is the diffusion coefficient in the phase space of Cartesian coordinates of crystal lattice, $D_r = D_r(r, t)$; $\alpha$ are connected with physical parameters of the problem: $D_r = D_{r0} \cdot \left( 1 + \alpha \left( \langle r^2 \rangle - \langle r \rangle^2 \right) \right)$.

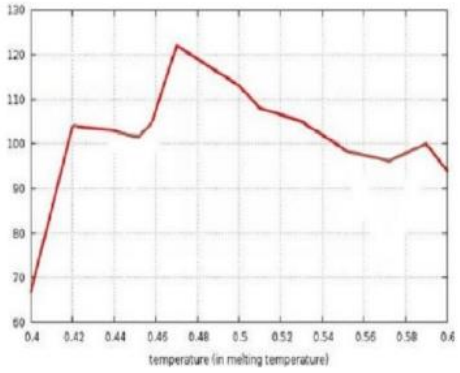
## RESULTS AND DISCUSSION

Calculation of *f*(*g*, *x*, *y*, *z*, *t*) allows us to find the dependence of the number of pores in structures of the sample (Fig. 1) on their sizes along the *Z* axis (distance from

the irradiated surface). In Fig. 3a two histograms are presented for two moments of time: abscissa axis presents the size structures (in the lattice parameters), ordinate axis presents the number of pores in the structure for the $4keV\ Xe^{++}$ ion flux and the dose is $10^{16}cm^{-2}$ for the layer temperature 1532 $K$. A structure consisting of two pores is taken into account if the distance between the centers of mass of gas inclusions is less than or equal to the sum of their radii plus the lattice parameter of the material. Porosity is used to estimate stresses in thin layers and at their boundary. Using the VGDs distributions (pore sizes and their coordinates), we can calculate the average length of defects in the lattice of a SiC / Mo sample for planes parallel and perpendicular to the flow (Fig. 3b and c).




a




b

Fig. 3a. Histograms of the dependence of the number pores in structures per unit volume of the silicon carbide layer on their size (projection on the Z-axis in the lattice parameters).

Fig. 3b. Dependence of the maximum length of structures of porosity related to the $Xe^{++}$ flux direction to the silicon carbide sample on $T/T_{melt}$.

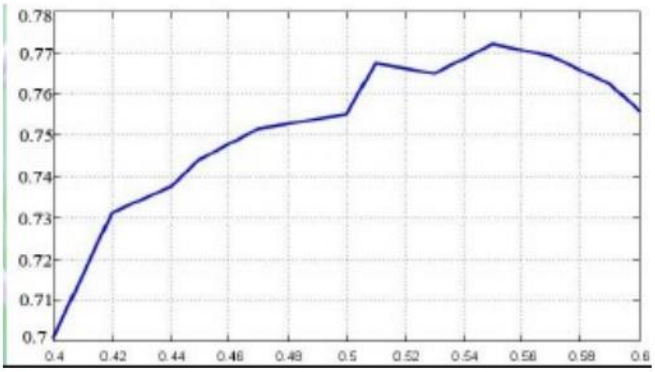



Fig. 3c. Dependence of the maximum length of structures of porosity related to the $Xe^{++}$ flux perpendicular direction to the silicon carbide sample on $T/T_{melt}$. The ordinate of the graph shows the average length of the pore structures in fractions of the size along the x axis, along the abscissa of the temperature, where the melting point of silicon carbide is (the end of calculations is 1.5 ms).

Calculate the average sizes of pore structures in a volume of 1x1x0.5 μm of 3C-SiC material depending on the temperature of the sample (a series of calculations is given under the same irradiation conditions) are shown in Fig. 4. Porosity Fig. 5 is used to estimate in thin layers and at their boundary.

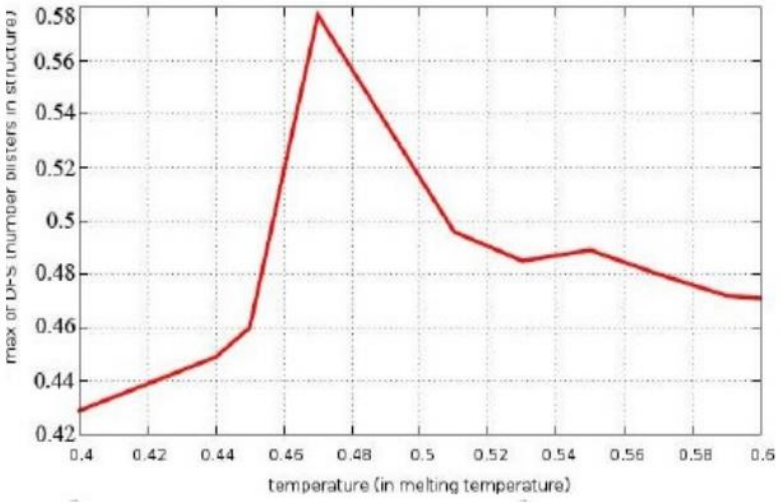



Fig. 4. Dependence of the most probable number of blisters in extended structures (the number of in extended structures is related to the total the number of VGDs in the sample)in dependence on temperature in fractions of temperature melting of the least refractory material 3C-SiC / Mo.

Modeling of implantation of the ions *Xe*++ showed that during the phase

transition the distribution of pore sizes evolves via the non-equilibrium stages. Consideration of Brownian motion of defects reveals the structure of defects arising under the action of $U(x,y,z)$ . In Fig 3b a number of structure defects per unit volume of SiC along the gas flow direction is presented as a function of temperature. Spherical pores of different diameters form specific structures during Brownian diffusion. The total length of structures is less than 30 lattice parameters of *3C-SiC*, which is used in assessing the degree of dispersion of the medium with inclusions of porosity and of amorphization.

Non-stationary kinetic equation being a 2-nd order partial differential equation with nonlinear coefficients is solved numerically within the method of stochastic molecular dynamics. Non-equilibrium size and spatial distributions of nuclei corresponding to an initial stage of a phase transition of the 1-st type in a bulk are obtained. Both, condensation of silicon carbide vapor in the discharge plasma, and structure of gas pores in the crystal lattice of the surface irradiated by the inert gas ions are described by quasilinear stochastic Ito equations in the sense of Stratonovich. Distribution functions for the size pores are calculated similar to [6] for the melt droplets (nano powders). The porosity in thin films are calculated using $10^6$ trajectories of random processes of the model.

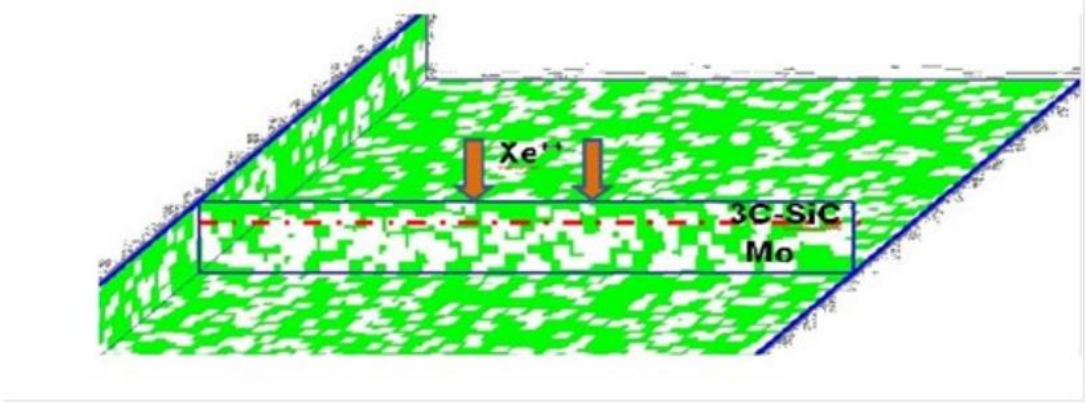
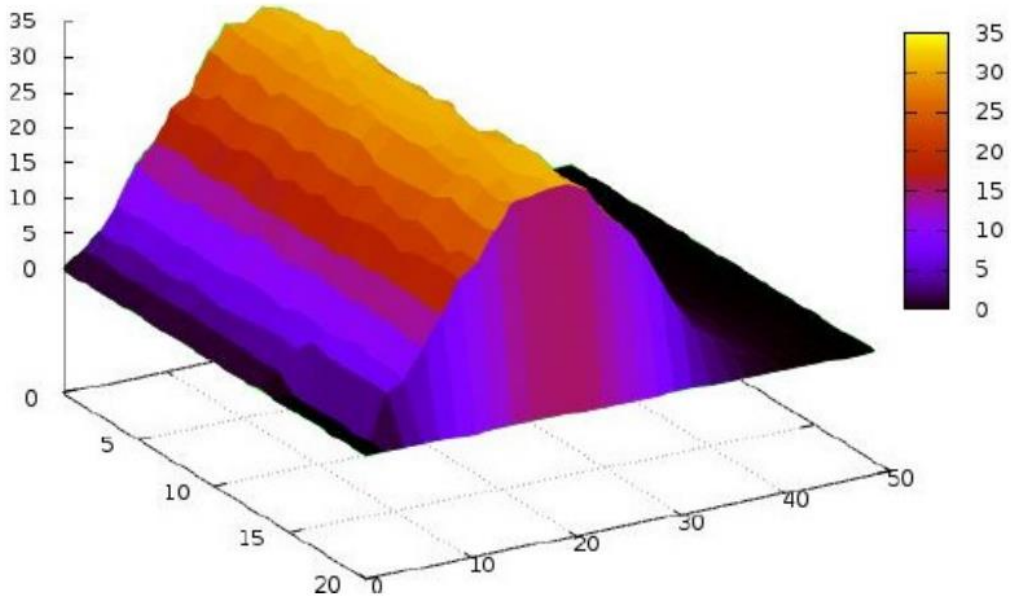



Fig. 5: The projections of the porosity(white color) on the section planes of the irradiated two layers sample(Fig.1a).




a

Fig.6a: Initial state DF for Brownian motion calculation. DF postponed on axis Z and counted from exposured surface, $f(|r_i - r_j|, Z)$, i,j=1,…,N, N is number VGDs corresponding to concentration $N_C$ in the sample (see Fig.1a, 3C-SiC layer).

Defect concentration Fig.6a is $N_c = 10^2$ cm$^{-3}$. Temperature sample is 1500K,radiation dose of $Xe^{++}$ is $10^{16}$cm-$^{2}$.

Dependence of the DF VGDs (arb. units, Z axis) on the distances between blisters (in the 3C-SiC lattice parameters) and on the distance from the irradiated surface at various concentrations (b, c, d). One division along the X axis is 30 lattice parameters. One division along the Y axis is 3.8 lattice parameters Defect concentration $N_c = 10^2$ cm$^{-3}$.

b

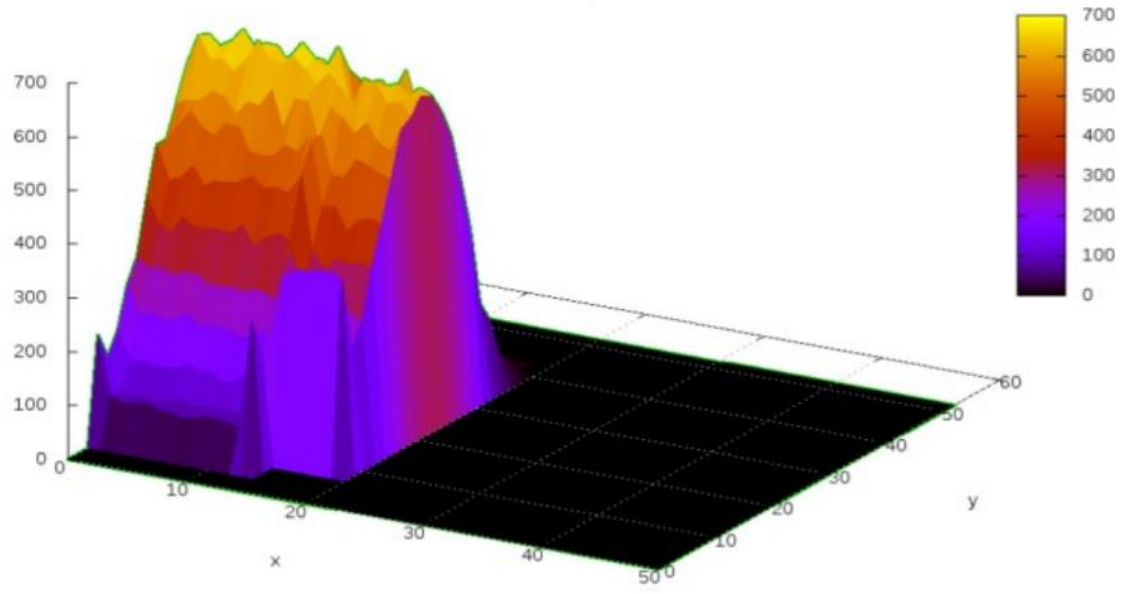



Fig.6 (b). The same that (a) in final state of run.

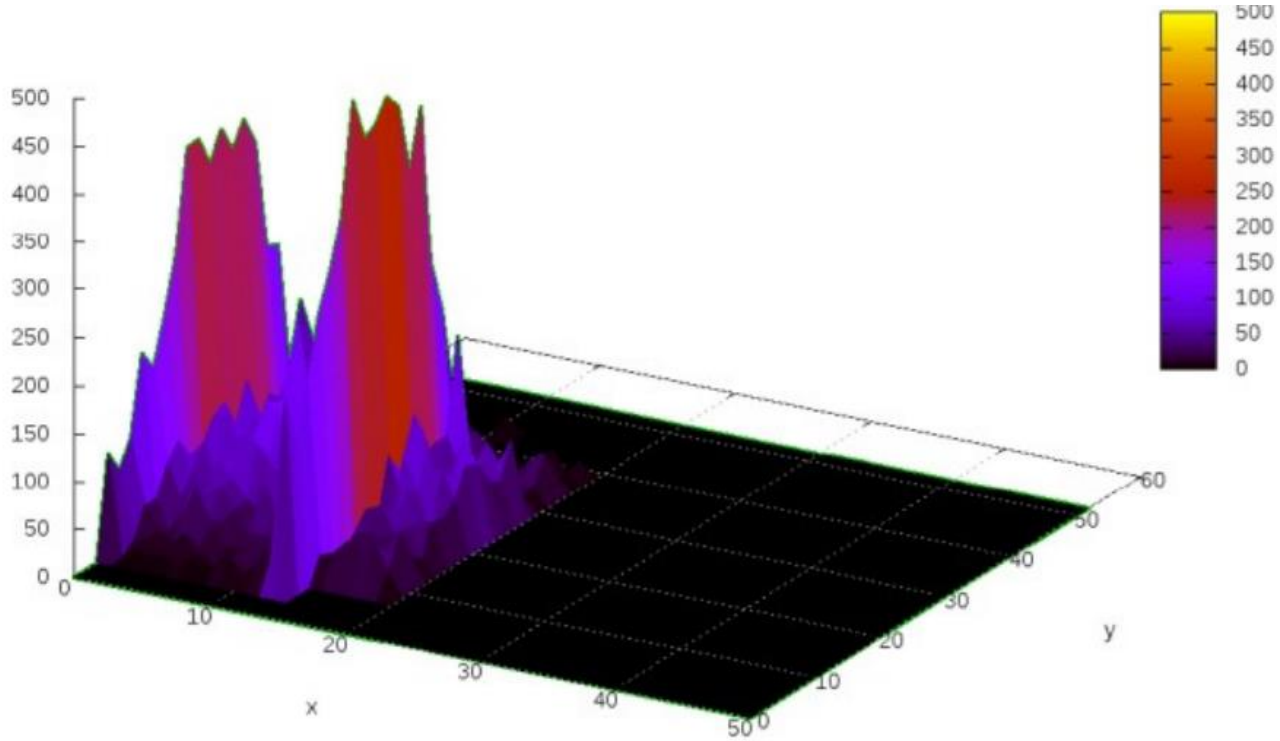
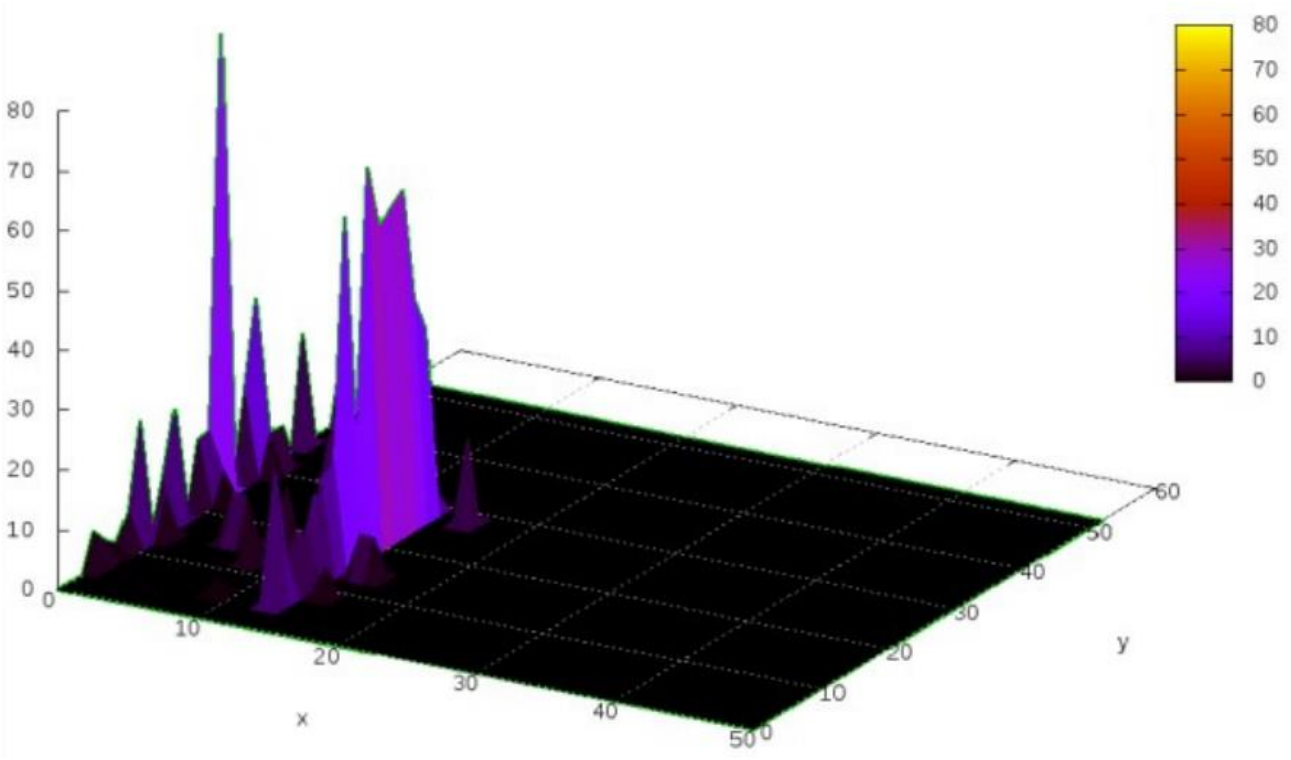



Fig.6 (c). The same that (a) at $T_{fin}$, $N_c = 10^3$ $cm^{-3}$.




d

Fig.6 (d). The same that (a) at $T_{fin}$, $N_c = 10^4$ $cm^{-3}$.

The simulation of the nonequilibrium stage of the phase transition at the initial nonequilibrium stage presented in the work continues the analysis of damages in the

crystal lattice, which are analyzed from the point of view of the appearance of porosity structures. The approach of blisters during their movement under the action of the potential of elastic forces occurs at times less than 100 microseconds, the potential of indirect elastic interaction is collective because it is calculated in a self-consistent manner. The acoustic phonons of the lattice during the formation of defects change the character of vibrations, which was taken into account when deriving the calculation formulas, the change in the coordinates of the pores changes the potential, which noticeably affects the motion of non-point defects and the concentration of defects also changes the length of the porosity structures. After calculating the formation of damage at different temperatures, it was found that the length of the structures is different in the direction along the direction of introduction of ions of inert times and perpendicular to it, the maximum length is reached at different temperatures, nonlinearly depending on it.

The formation of spatial-temporal structures in the phase space of stochastic dynamic variables provides new information on the behavior of the model in an open physical system and expands the understanding of the kinetics of phase transitions at a fluctuatingly unstable stage and complements the model of damage to the crystal lattice by a numerical analysis of the formation of non-point defects.

## ACKNOWLEDGMENTS

The author is grateful to the RFBR for partial support of the work by grant 18-01-00436 and collegues A.L. Bondareva and T.A. Averina for many years of cooperation in modifying the numerical algorithm and in visualizing the results and also to Yuri Gorbachev for the head of the Fifth mini-symposium “Physical-chemical gas-dynamics: Non-equilibrium processes modeling and simulation” at International Conference of Numerical Analysis and Applied Mathematics ICNAAM 2019, Rodos, Greece, September 23-28, 2019 for useful discussions of her presentation at this symposium.

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