

UDC 535.326:539.05:539.1.074.4:621.385.6

DOI <https://doi.org/10.32782/pet-2023-1-6>

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To cite this article: Trokhimchuck, P. (2023). Do pytannia pro pryrodu ta modeliuvannia zarodkoutvorennia ta krystalizatsii [To question about nature and modeling the processes of nucleation and crystallization]. *Physics and Education Technology*, 1, 40–46, doi: <https://doi.org/10.32782/pet-2023-1-6>

TO QUESTION ABOUT NATURE AND MODELLING THE PROCESSES OF NUCLEATION AND CRYSTALLIZATION

The main problems of nature and modelling the nucleation and crystallization are discussed. Two aspects of this problem: thermodynamical and electrodynamic, are discussed. Nucleation processes are classed as heterogeneous or homogeneous. Main thermodynamic theories and models of thermodynamic nucleation and crystallization, including Stranski-Krastanow model, are analyzed. It is shown, that these theories are explained the classic crystallization processes and methods, including Kiropoulos method, Chochralsky method, Bridgman-Stockbarger method, methods of zonal crystallization, growing with solution mixture. Vitaliy Stafeev electrostatic phason model, cascade theories of excitation of corresponding chemical bonds (coordination numbers) are represented electromagnetic models. Vitaliy Stafeev model allow to estimate minimal sizes of new phases – phasons and may be used for continuous and pulse regimes of nucleation and crystallizations. Cascade theories allow to explain the laser-induced pulses phase transformations. In this case we can have new phases with an increase in the degree of order (irradiation of the unstable or metastable structures), as well as with a decrease in the degree of order (irradiation of the stable structures). Examples of the use of these theories to describe phase changes during laser irradiation of silicon, germanium, indium antimonide, and indium arsenide are given. These theories may be used for all possible matter: from neorganic to biological. The problem of saturation is on of central problem of these theories. This problem has two ways of resolution. For thermodynamical theories and models it is saturation of solutions and dynamic of change this saturation. For electromagnetic theories and models it is the saturation of excitation and intensity of this excitation. The perspective of development and application these methods are discussed.

Key words: nucleation, crystallization, V. Stafeev model, Stranski-Krastanow model, saturation, Relaxed Optics, cascade models, modeling.

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Бібліографічний опис статті. Трохимчук, П. (2023). До питання про природу та моделювання зародкоутворення та кристалізації. *Фізика та освітні технології*, 1, 40–46, doi: <https://doi.org/10.32782/pet-2023-1-6>

ДО ПИТАННЯ ПРО ПРИРОДУ ТА МОДЕЛЮВАННЯ ПРОЦЕСІВ ЗАРОДКОУТВОРЕННЯ ТА КРИСТАЛІЗАЦІЇ

Обговорюються основні проблеми природи та моделювання зародкоутворення та кристалізації. Обговорюються два аспекти цієї проблеми: термодинамічний і електродинамічний. Процеси зародження класифікуються як гетерогенні або гомогенні. Проаналізовано основні термодинамічні теорії та моделі термодинамічного зародкоутворення та кристалізації, включаючи модель Странського-Крастанова. Показано, що ці теорії пояснюють класичні процеси та методи кристалізації, включаючи метод Кіропулоса, метод Чохральського, метод Бріджмена-Стокбаргера, методи зонної кристалізації, вирощування кристалів з рідких розчинів. Електромагнітні моделі представлені електростатичною фазною моделлю Віталія Стафеева,

каскадними теоріями збудження відповідних хімічних зв'язків (координаційних чисел). Модель Віталія Стафєєва дозволяє оцінити мінімальні розміри нових фаз – фазонів і може бути використана для неперервного та імпульсного режимів зародкоутворення та кристалізації. Каскадні теорії дозволяють пояснити фазові перетворення під впливом лазерних імпульсів. У цьому випадку ми можемо мати нові фази як зі збільшенням ступеня впорядкованості (опромінення нестабільних або метастабільних структур), так і зі зниженням ступеня впорядкованості (опромінення стабільних структур). Наведено приклади використання цих теорій для опису фазових змін під час лазерного опромінення кремнію, германію, антимоніду індію та арсеніду індію. Ці теорії можна використовувати для всіх можливих середовищ: від неорганічних до біологічних. Проблема насичення є однією з центральних проблем цих теорій. Ця проблема має два шляхи вирішення. Для термодинамічних теорій і моделей це насичення розчинів і динаміка зміни цього насичення. Для електромагнітних теорій і моделей це насиченість збудження та інтенсивність цього збудження. Обговорено перспективи розвитку та застосування цих методів.

Ключові слова: зародкоутворення, кристалізація, модель В. Стафєєва, модель Странського-Крастанова, насичення, релаксаційна оптика, каскадні моделі, моделювання.

INTRODUCTION

In modern Physical Chemistry the **nucleation**, the initial process that occurs in the formation of a crystal from a solution, a liquid, or a vapour, in which a small number of ions, atoms, or molecules become arranged in a pattern characteristic of a crystalline solid, forming a site upon which additional particles are deposited as the crystal grows (Markov, 2017). That is why thermodynamic theories of nucleation and crystal growth were initially developed.

In this work, we will analyze the theories and models that are successfully used and can be used in the creation of new technologies for the miniaturization of elements of optoelectronic systems.

A typical representative of such a theory is the Stransky-Krastanow theory (Bauer, 1958; Stranski, 1938). These theories qualitatively explain the results of the main methods of growing crystals: melt, solution and gas transport methods.

However, with the development of micro and nanotechnologies, the problem of estimating the minimum sizes of the formed new phases has become relevant. That is why Vitaly Stafeev's theory of phasons appeared (Stafeev, 2005).

With the development of laser technologies, it became necessary to create models and theories of nucleation and crystallization that take into account the irradiation regimes. This is how cascade models of laser-generated phase transformations appeared (Trokhimchuck, 2016).

For example, a two-dimensional crystal lattice was used for indium antimonide and indium arsenide. The cascade model of the excitation of the corresponding chemical bonds in the excitation saturation mode made it possible to explain the laser-induced defect formation in these materials (Trokhimchuck, 2016).

The cascade model, which was built for silicon and germanium based on their phase diagram, where coordination numbers were taken into account, made it possible to explain the laser-induced cascades of phase transformations with a decrease in the order of symmetry of the crystal lattice and to explain the corresponding experimental results (Trokhimchuck, 2016).

STRANSKI-KRASTANOV MODEL

Stranski-Krastanow growth (Stranski, 1938) is one of the three primary modes by which thin films grow epitaxially a crystal surface or interface. Also known as 'layer-plus-island growth', the SK mode follows a two step process: initially, complete films of adsorbates, up to several monolayers thick, grow in a layer-by-layer fashion on a crystal substrate. Beyond a critical layer thickness, which depends on strain and the chemical potential of the deposited film, growth continues through the nucleation and coalescence of adsorbate 'islands'. Volmer-Weber, and Frank-van der Merwe mechanisms were systematically classified as the primary thin-film growth processes (Bauer, 1958; Markov, 2017). Since then, SK growth has been the subject of intense investigation, not only to better understand the complex thermodynamics and kinetics at the core of thin-film formation, but also as a route to fabricating novel nanostructures for application in the microelectronics industry.

The growth of epitaxial (homogeneous or heterogeneous) thin films on a single crystal surface depends critically on the interaction strength between adatoms and the surface. While it is possible to grow epilayers from a liquid solution, most epitaxial growth occurs via a vapor phase technique such as molecular beam epitaxy (MBE). In Volmer-Weber (VW) growth, adatom-adatom interactions are stronger than those of the adatom with the surface, leading to the formation of three-di-

mensional adatom clusters or islands. Growth of these clusters, along with coarsening, will cause rough multi-layer films to grow on the substrate surface. Antithetically, during Frank–van der Merwe (FM) growth, adatoms attach preferentially to surface sites resulting in atomically smooth, fully formed layers. This layer-by-layer growth is two-dimensional, indicating that complete films form prior to growth of subsequent layers (Bauer, 1958; Markov, 2017). Stranski–Krastanow growth is an intermediary process characterized by both 2D layer and 3D island growth. Transition from the layer-by-layer to island-based growth occurs at a critical layer thickness which is highly dependent on the chemical and physical properties, such as surface energies and lattice parameters, of the substrate and film (Bauer, 1958; Markov, 2017). Fig. 1 is a schematic representation of the three main growth modes for various surface coverages.

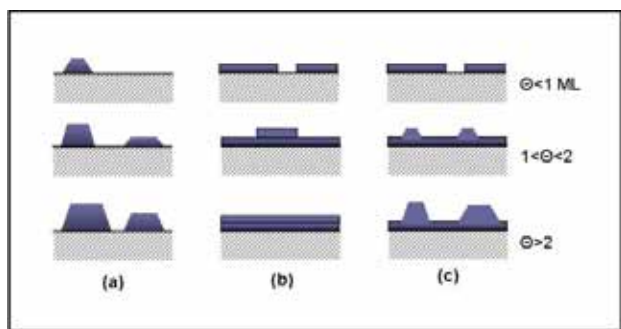


Fig. 1. Cross-section views of the three primary modes of thin-film growth including (a) Volmer–Weber (VW: island formation), (b) Frank–van der Merwe (FM: layer-by-layer), and (c) Stranski–Krastanov (SK: layer-plus-island). Each mode is shown for several different amounts of surface coverage, Θ

Determining the mechanism by which a thin film grows requires consideration of the chemical potentials of the first few deposited layers (Bauer, 1958; Markov, 2017). A model for the layer chemical potential per atom has been proposed by Markov (Markov, 2017) as:

$$\mu(n) = \mu_{\infty} + [\varphi_a - \varphi'_a(n) + \varepsilon_d(n) + \varepsilon_e(n)], \quad (1)$$

where μ_{∞} is the bulk chemical potential of the adsorbate material, φ_a is the desorption energy of an adsorbate atom from a wetting layer of the same material, $\varphi'_a(n)$ the desorption energy of an adsorbate atom from the substrate, $\varepsilon_d(n)$ is the per

atom misfit dislocation energy, and $\varepsilon_e(n)$ the per atom homogeneous strain energy. In general, the values of φ_a , $\varphi'_a(n)$, $\varepsilon_d(n)$, and $\varepsilon_e(n)$ depend in a complex way on the thickness of the growing layers and lattice misfit between the substrate and adsorbate film.

In the limit of small strains, $\varepsilon_d(n) \ll \mu_{\infty}$, the criterion for a film growth mode is dependent on $\frac{d\mu}{dn}$.

- VW growth: $\frac{d\mu}{dn} < 0$ (adatom cohesive force

is stronger than surface adhesive force), (2)

- FM growth: $\frac{d\mu}{dn} > 0$ (surface adhesive force

is stronger than adatom cohesive force). (3)

SK growth can be described by both of these inequalities. While initial film growth follows an FM mechanism, i.e. positive differential μ , non-trivial amounts of strain energy accumulate in the deposited layers. At a critical thickness, this strain induces a sign reversal in the chemical potential, i.e. negative differential μ , leading to a switch in the growth mode. At this point it is energetically favorable to nucleate islands and further growth occurs by a VW type mechanism (Bauer, 1958). A thermodynamic criterion for layer growth similar to the one presented above can be obtained using a force balance of surface tensions and contact angle.

STAFEEV PHASON MODEL

The problem of the creation new phase centers (phasons) is represented in Stafeev model (Stafeev, 2005). It is electrostatic theory.

Experimental data (Stafeev, 2005) shown that phasons are mono dispersive particles. Its sizes are depended from properties of its matter and environment of its formation. New condensed phase centers may be generated under vaporation, on substrate, in gas or liquid environment and directly in solid state. Under change of chemical compound phasons change its sizes in jumping way. For various matters minimal sizes of phasons are changed from 1 to 20 nm (Stafeev, 2005). Nanoparticles with these sizes are observed in soli state. It is structural defects – clusters (Stafeev, 2005). In some environment crystallization centers are covered by shell, its form stands more lock-in. Its nanoparticles are analogous to special “atoms” – small bricks of new matter (Stafeev, 2005). It may be included in solution, including solid, and created own condensed matter. For objects of little sizes, classical thermodynamical approaches and charac-

teristics (surface energy a.o.) cannot be used. However, must be existed some physical characteristics of matter and environment, which are determined minimal sizes of phasons.

As rule take into account, that nanoparticles are neutral (Stafeev, 2005). But investigations in region of phase transitions were shown that new phase centers (phasons) must be have electrical charge. For example (Stafeev, 2005), between growth ice layer from water may be generated sufficiently great (decathlons and hundreds Volts) difference of electrical potentials. Under phase transitions in liquid crystals great difference of electrical potentials is observed (Stafeev, 2005).

Atoms is determined the properties of chemical elements, molecules – chemical properties of substance. Must be existed structural unit that determine physical and other properties of condensed phases of substance. Last is depended not only from state of substrate but from its phase. Phasons may be completed from comparatively small numbers of molecules. Some quantitative threshold must be existed. Beginning from this threshold molecular cluster is transformed to thermodynamically stable phasons (Stafeev, 2005).

One of the important parameter of any phase is electrochemical potential. Therefore on its border with another phase junction difference of electrical potentials, which is stipulated of difference of electrochemical potentials, must be generated. It may be realized only in presence in each with contact phase's electrical charges with opposite signs. Cluster of bound molecules may be thermodynamically stable center of new phase after formation of proper junction difference of potentials only.

Minimal size of phason is determined from condition, that unit electrical charge q in environment with electrical constant ε on its capacity c is provided the generation of necessary junction difference of potentials $\varphi = q/\varepsilon c$. Center of new phase with minimal sizes was called phason (Stafeev, 2005). These centers may be having various forms. For simplification of further analysis we allow that center of new phase is spherical capacitor with diameter d_0 . For this suggestion (Stafeev, 2005):

size of phason – $d_0 = 3,6/\varepsilon\varphi, nm$;

number molecules in phason – $n_f = 10^2 \pi \rho d_0^3 / M$;

mobility – $\mu = \varepsilon\varphi/6\pi\eta = 5,3 \times 10^{-7} \varepsilon\varphi/\eta, cm^2/V \times s$;

diffusive coefficient – $D = 4,6 \times 10^{-11} \varepsilon\varphi T/\eta, cm^2/s$,

where φ in Volts; q in Coulomb; η viscosity in Poissons; N – Avogadro number; T – temperature; M – molecular weight; ρ is density of phason substrate in g/cm^3 .

Diameter, mobility and diffusive coefficient of phasons are determined of electric constant of surrounding environment and difference of electrochemical potentials.

Sign of difference of electrochemical potentials is determined the sign of charge of center of new phase. For vacuum and gas, the role of difference of electrochemical potentials has energy of chemical affinity to electron (or proton) atoms and molecules of phason substrate w . For positive sign of affinity to electron phason charge will be negative, for positive to proton – positive. Knowledge of phason size allow estimating the affinity to electron (proton) its substance or difference of electrochemical potentials phason and substance of basic phase.

Phasons may be creating more large clusters – polyphasons. It must be quantized on sizes $d = nd_0$, where $n = 1, 2, 3 \dots$

CASCADE MODELS OF STEP-ON-STEP SATURATED EXCITATION OF CORRESPONDING CHEMICAL BONDS

For modeling in cascade model were used two-dimensional picture the crystal lattice A_3B_5 (Fig. 1 a) (Trokhimchuck, 2016) and phase diagram of Si (Fig. 1 b) (Trokhimchuck, 2016).

The profiles of the distribution the photostimulated donor centers in subsurface layers $InSb$ and $InAs$ are may be have various form (Trokhimchuck, 2016).

The samples of p-type conductivity are irradiated. For intensity of irradiation $I_0 > 0,01 J \cdot cm^{-2}$ for $InSb$ and $I_0 > 0,012 J \cdot cm^{-2}$ for $InAs$ the n-layers on p-type materials are created. For intensity of irradiation $I_0 < 0,1 J \cdot cm^{-2}$ for $InSb$ and $I_0 < 0,16 J \cdot cm^{-2}$ for $InAs$ the profiles of the distribution of donor centers are represented the Buger-Lambert law (law of absorption the light in homogeneous media). For further increasing the irradiated intensity the profiles of the concentration donor centers have diffusion nature. The visible destruction of the irradiated semiconductor melting, the change of the surface color) had place for $I_0 > 0,3 J \cdot cm^{-2}$ for $InSb$ and $I_0 > 0,5 J \cdot cm^{-2}$ for $InAs$. This effect has oriental character (Trokhimchuck, 2016). For crystallographic direction $\{111\}$ the process of the creation damages is more effective as for direction $\{110\}$.

Analogous results were received for the indium arsenide too. Time and energy characteristics of these processes on may be estimated with the help of next way. In further we'll use two-dimensional representation of crystal lattice *InSb* (Fig.7). Bond 1 is corresponded to band gap and has value 0,18 eV, bond 2 – 1,95 eV and bond 3 – 2,15 eV (Trokhimchuck, 2016). For *InAs* bond 1 has value 0,36 eV, bond 2 – 3,8 eV and bond 3 – 4,2 eV (Trokhimchuck, 2016).

For these crystals the energy of these bonds are equaled the energy of band gap E_g (0,18 eV for *InSb* and 0,36 eV for *InAs* at room temperature). On Fig. 2 a) this bond is signed as 1. For this bond ions *In* and *Sb* (or *In* and *As*) are placed on minimal distance (the sum of proper covalent radiuses). Other chemical bonds in this crystal symmetry have more long sizes. With geometrical point of view in crystal direction {111} the cross section of effective interaction the light quantum with bond 1 is more effective than for direction {110}. The angle among bond 1 and direction {110} is 37,5°. Quanta of ruby laser in linear regime of the irradiation are not interacted with another bonds practically because it energies are less than energy of this bond. The correlation of effective square of bond 1 for directions {110} and {111} is explained the proper experimental data (Trokhimchuck, 2016).

Straight method of the estimation the energetic characteristics this processes may be realized in the next way. Energy of “disruption” of chemical bonds of one type is equalled

$$E_{di} = N_i E_i, \quad (4)$$

where N_i – a density of proper bonds; E_i – energy of a disruption (ionization) one bond.

For the *InSb* $N_1 = N_2 = N_3 = \frac{N_0}{2}$ and are equaled $1.4 \cdot 10^{22} \text{ cm}^{-3}$, $E_1 = E_g = 0.18 \text{ eV}$ and therefore $E_{d1} = N_1 E_g = 403.2 \text{ J/cm}^3$ and $E_{d2} = N_2 E_2 = 4368 \text{ J/cm}^3$. Surface density of irradiation may be determined with the help of next formula (Trokhimchuck, 2016)

$$E_{sis} = \frac{E_{di}}{\alpha_i}, \quad (5)$$

where α_i – proper absorption factor, for the first bonds of *InSb* $\alpha_1 = 2 \cdot 10^5 \text{ cm}^{-1}$, for second – $\alpha_2 \sim 10^5 \text{ cm}^{-1}$. Second absorption factor is nonlinear and take into account the effect of blooming. For *InSb* these values are next $E_{s1s} = 0.002 \text{ J/cm}^2$ and $E_{s2s} = 0.04368 \text{ J/cm}^2$. These values must be multiplied on 2 (with including reflection) and therefore real values are next $E_{s1sr} = 0.004 \text{ J/cm}^2$, $E_{s2sr} = 0.08797 \text{ J/cm}^2$ and $E_{sr(2)} \approx 0.092 \text{ J/cm}^2$. Energy of “disruption” of third chemical bonds (Fig.4.4) is equalled $E_{d3} = N_3 E_3 = 4816 \text{ J/cm}^3$. If $\alpha_3 \sim 10^5 \text{ cm}^{-1}$ we have $E_{s2s} = 0.04816 \text{ J/cm}^2$ and $E_{s3sr} = 0.096 \text{ J/cm}^2$. Summary surface density of energy of three bonds is equalled $E_{sr(3)} \approx 0.188 \text{ J/cm}^2$. Value $E_{sr(2)} \approx 0.092 \text{ J/cm}^2$ is represented of maximal distribution n-centers in subsurface region

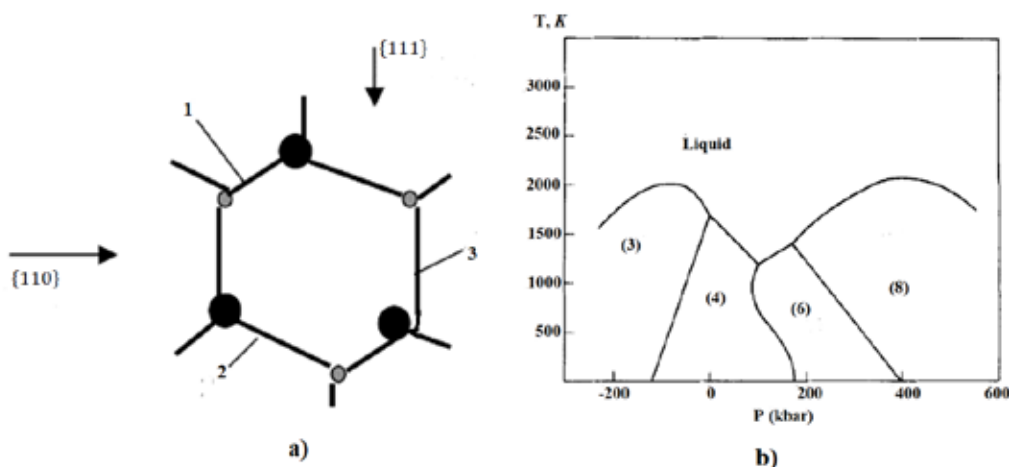


Fig. 2. a) Two-dimensional picture the crystal lattice A_3B_5 (including *InSb* and *InAs*) the cubic symmetry. Bond 1 is pure covalent; b) a schematic phase diagram for *Si(CN)*. The coordination numbers (CN) of the various phases are indicated

and $E_{\Sigma r(3)} \approx 0.188 J/cm^2$ – “thermal diffusive” case (Trokhimchuck, 2016).

The fact is that this radiation leads to phase modifications of the pure material. The use of CO₂ laser radiation leads to annealing of defects and crystallization of ion-implanted layers. Moreover, the irradiation modes (impulse or stationary) do not matter, the main role is played by the integral dose. That is, in this case, we have photochemical processes (Trokhimchuck, 2016).

Now we show the using of cascade model for the explanation experimental data of laser-induced phase transformations in silicon, germanium, carbon and titanium. It was called as case the structural phases (Trokhimchuck, 2016).

The question about the influence of saturation of excitation on effects of RO may be represented as process of transitions between stable and metastable phases too. Now we'll estimate the influence of parameters of irradiation (including spectral) on irreversible changes and transformations in *Si* and *Ge*. Spectral dependences of absorbance of various structural modification of *Si* are represented in (Trokhimchuck, 2016). Now we'll be estimated intensities of eximer, Ruby and Neodymium laser irradiation (wavelengths of irradiation are 0,248 μm , 0,69 μm and 1,06 μm properly of silicon and germanium, which are necessary for the creation of proper irreversible changes in irradiated semiconductor. As shown in (Trokhimchuck, 2016), absorbance of the Neodymium laser radiation in silicon is equaled 100 cm^{-1} , second harmonic of Neodymium laser – 10⁴ cm^{-1} , eximer laser – 10⁶ cm^{-1} .

Crystal semiconductors *Si* and *Ge* have, basically, the structure of diamond. Volume atomic density of elementary lattices may estimate according to formula [19]

$$N_a = \frac{\rho N}{A}, \quad (6)$$

where ρ – density of semiconductor, N – Avogadro number, A – a weight of one gram-atom. For *Si* $N_{aSi} = 5 \cdot 10^{22} cm^{-3}$, and for *Ge* $N_{aGe} = 4.4 \cdot 10^{22} cm^{-3}$.

But *Si* and *Ge* may be crystallized in lattices with hexagonal, cubic, trigonal and monoclinic symmetry. Phase diagram of *Si* as function of coordination number is represented on Fig. 1 b) (Trokhimchuck, 2016).

Coordination number (CN) 8 is corresponded of diamond lattice, CN 6 – hexagonal lattice, (CN) 4 and (CN) 3 – other two lattices (trigonal and monocline). It should be noted that melting temperatures of these phases are various. Volume density of CN is equaled $CN \cdot N_a$. For diamond symmetry of lattice this value is $8N_a$. In other words, a change in the coordination number is not necessarily related to a change in type of crystal syngonia. Thus, the coordination numbers 8 and 6 can correspond to two different cubic symmetries, and at the same time, CN 6 corresponds to hexagonal syngonia (Boky, 1971), which corresponds to the corresponding experimental data (Trokhimchuck, 2016).

Roughly speaking, transition from one phase to another for regime of saturation of excitation may be modeled as one-time breakage of proper numbers of chemical bonds, which are corresponded to the difference of CN of proper phases. For example, two bonds breakage is caused the phase transition from diamond to hexagonal structure. One bond breakage in the regime of saturation is caused to generation of laser radiation.

Results of calculation of volume and surface densities of energy, which are necessary for breakage of proper number of bonds in regime of saturation of excitation, are represented in (Trokhimchuck, 2016) and they allow us to explain the corresponding experimental data.

CONCLUSIONS

1. Stranski-Krastanow thermodynamic model of nucleation and crystallization is analyzed.

2. Electrostatic Stafeev phason theory is represented. This theory allow to estimate minimal sizes of phasons.

3. Cascade models of excitation the corresponding chemical bonds or coordination numbers in the regime of saturation allow to explain laser-induced phase transformations for various regimes of irradiation.

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