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Hetero- and low-dimensional structures

Self-organized structures induced by external white noise and nanosized levels of their formation in the non-crystalline As-S(Se) semiconductor systems

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Abstract. Discussed in this paper are the singularity and self-organizing effect of instability and randomness under the influence of external white noise on formation of non-crystalline materials. The random nature of the receiving medium together with the disorganizing effect was found to be capable to initiate formation of qualitatively new self-organized structures in non-crystalline solids. Also analyzed in the paper is the effect of a random temperature field applied to the melt during the cooling process in non-crystalline As-S(Se) semiconductor systems. The conditions for a non-crystalline system in a fluctuating external environment to adjust its properties to the average properties of the environment and to correspond to the deterministic case were identified. Furthermore, the conditions for non-additive reaction of the system to a random environment and formation of a new mode of energy conversion in the self-organized structure at the nanoscale level are determined. The spectrum of the structures created in this way is more diverse as compared to the spectrum corresponding to respective deterministic conditions.

Keywords: bifurcation diagram, fractality, nanosized effects, synergetics, white noise, non-crystalline semiconductors, self-organized structures.

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1. Introduction

Amorphous substances are widely used in modern semiconductor devices along with crystalline materials. A substantial advantage of amorphous materials over crystalline ones is their processability (relative simplicity and lower energy consumption of technological processes) as well as capability to change the composition and properties [1–3]. Non-crystalline materials enable creation of devices that are easy to microminiaturize. Moreover, they can be easily integrated with crystals in the same device [3]. Development of intelligent nano-environments is also an important incentive for advancing research on non-crystalline and smart materials, in particular, for artificial intelligence applications [4–6]. Non-crystalline chalcogenides of the As-S(Se) system are actively studied due to their unique properties. These properties, together with the developed methods of obtaining thin films provide new opportunities for practical application of such materials [7]. In particular, mentioned applications include optical memory disks, integrated circuits on thin flexible substrates, high-speed photosensitive drums for photo-copying, photoresist materials for lithography, optical elements of laser technology, holograms and elements of integrated optics, highly sensitive thermoelectric converters and photocells, switches, and photoelectric materials [5, 7–10]. To ensure reproducibility of the results obtained for amorphous film structures, the main parameters of the film synthesis must be thoroughly controlled. Therefore, the most significant characteristics of the stochastic variability of the film formation conditions must be accounted for in the technological processes.

Study of self-organization processes in non-crystalline systems in fluctuating non-equilibrium environments implies revision of the views on the randomness effect [6]. Randomness plays an important constructive role in the theory of self-organization. The issue of the influence of external noise on open systems is constantly addressed [8, 11] mainly due to the fact that the parameters of a macroscopic system (including the bifurcation parameters) are externally oriented and subject to fluctuations. These fluctuations are perceived by the system as external noise. In the studies of the impact of external noise on *e.g.* cooling rate, irradiation power and bioactive external fields, this impact on non-crystalline solids is averaged. This averaging provides an integral effect, and the macroscopic system "adjusts" its state to the averaged conditions of the medium. In fact, stochastic variability of the fabrication conditions in technological media leads to the fuzziness of the system states, which approaches some average value [10, 11]. The assumption of ultimately secondary role of fluctuations is based on a linear-type relationship between the system and the environment. However, theoretical and experimental studies of melt cooling in fluctuating media with noise indicate the opposite, namely that the random nature of the medium together with the disorganizing effect can initiate formation of qualitatively new self-organized structures in non-crystalline solids [6]. The class of structures obtained in this way is qualitatively more diverse as compared to the possible spectrum corresponding to respective deterministic conditions. Hence, increase in stochastic variability of the receiving medium can determine the possibility of controlled structuring of non-crystalline materials depending on noise levels as well as the formation of nanoscale systems of self-organized structures. It can be highly relevant, in particular, in the development of smart non-crystalline materials [10, 11]. This aspect of the synergetic relationship between instability and randomness becomes evident considering functioning of information systems in non-crystalline semiconductor materials for artificial intelligence. It is reasonable since randomness and instability correlate with chaos-entropy, and system structuring and ordering are achieved through the flow of information – syntropy. These processes can be complementary during the formation of nanoscale self-organized structures in non-crystalline and smart materials under the influence of external noise.

This article is devoted to the topic outlined above. In particular, it studies self-organization processes of non-crystalline materials in the presence of external white noise. Our approach uses object-oriented modeling, which is *de facto* based on the principles of synergy. The article also considers the results of the study of the influence of cooling rate fluctuations in the white noise approximation on the formation of the nanoscale levels of self-organized structures in non-crystalline semiconductors of the As-S(Se) system.

2. External noise-induced transitions and their modeling

2.1. Research concepts and methods

Influence of external noise on the behavior of open systems has been widely discussed in the literature [6, 10, 12–14]. New and unexpected results have been obtained with the development and formation of synergy in this field. In particular, external noise has been found to cause not only a standard linear response in dynamic systems associated with fluctuations of their properties but also to determine a qualitative change in the system behavior. It follows therefore that the behavior of a highly non-equilibrium system can be defined not only by a deterministic external action but also by random fluctuations of environmental parameters interacting with the open system [15].

The following main aspects can be distinguished when modeling the problem of the influence of external random field on the behavior of macro- and microscopic characteristics of non-crystalline materials. Influence of the environment on a non-equilibrium system is described by the phenomenological equation containing an external random parameter. (The regularity of the changes in the medium during cooling or deposition can be set by using automated systems for controlling the external temperature field [10].) The quantities contained in the mentioned equation as well as the random stationary processes can be represented as follows:

$$q_t = q + \sigma_\xi \xi_t \,, \tag{1}$$

where q is the average cooling rate (corresponds to the deterministic case), ξ_t is the random external fluctuating field, and σ_ξ is the external noise intensity ($\sigma_\xi{}^2 = \langle \xi_t \cdot \xi_{t'} \rangle$, $\langle \cdots \rangle$ means averaging over the random field), respectively. As opposed to the fluctuations of the internal parameter of a non-crystalline system, which have microscopic origin, the stochasticity of the environment as a source of noise has a different nature, *i.e.*, the intensity of fluctuations in the medium can be controlled [16–18]. It is essential for the study of the influence of external noise on system behavior: $(dS)_i \geq 0$ is the entropy flux in the non-crystalline system and $(dS)_e < 0$ is the syntropy flux from the external environment [8], respectively. Moreover, it is possible to reduce the noise level, yet it is impossible to completely eliminate it, *i.e.* external noise never equals zero. While modeling external noise, the environment can be specified as a source of noise in accordance with the central limit theorem of the probability theory [15]. The stochastic differential equation (SDE) and the stochastic differential considering the external noise acquire the following forms [15]:

$$\frac{d\eta_\sigma(r,t)}{dt} = f_{q_t}\left(\eta_\sigma(r,t)\right), \quad d\eta_\sigma = f_{q_t}\left(\eta_\sigma\right) \cdot dt + G_q \cdot dW_t \,. \tag{2}$$

Here, $\eta_\sigma(r,t)$ is the ordering parameter of the non-crystalline system, $f_{q_t}\left(\eta_\sigma(r,t)\right) = h(\eta_\sigma) + q_t g(\eta_\sigma)$ is the nonlinear function describing local evolution of the component $\eta_\sigma(r,t)$ in space r and time t, $h(\eta_\sigma)$ and $g(\eta_\sigma)$ are the functions of $\eta_\sigma(r,t)$, W_t is the medium temperature field (thermostat), and G_q is the source of random noise, respectively. The change of the relative temperature of the dynamic instability of a non-crystalline solid can be used as a system order lines parameter:

$$\eta_\sigma = \left(T_m - T_{sg}\right) \big/ T_m \,,$$

which is directly determined experimentally and is related to the fraction of atoms in the soft atomic configurations [8]. In crystals, $T_{sg} = T_m$ and $\eta_\sigma = 0$.

In non-crystalline solids, $T_{sg} = T_{sg}(q) \le T_m$. The latter value decreases with the increase of q [8, 19]. Therefore, $0 \le \eta_\sigma \le 1$.

Taking into account Eq. (1), Eq. (2) may be presented as follows:

$$\frac{d\eta_\sigma(r,t)}{dt} = f_{q_t}(\eta_\sigma)dt + \sigma_\xi \cdot g(\eta_\sigma) \cdot dW_t = \\ = h(\eta_\sigma) + q_t \cdot g(\eta_\sigma) + \xi_t g(\eta_\sigma), \tag{3}$$

where $dW_t = \xi_t g(\eta_\sigma)$. The second summand in (3), $\approx \sigma_\xi g(\eta_\sigma)$ corresponds to the diffusion of the soft atomic configuration regions due to the influence of external noise.

Functional dependence between the random variables that model fluctuations of the environment is obtained as follows. We consider the approximation of external white noise with the state of the medium changing significantly faster than the macroscopic state of the system. We denote τ_m to be the characteristic time of the macroscopic evolution of the system or, identically, the time of the system relaxation to the stationary state η_{σ_s} :

$$\tau_m = \left|1/\omega(\eta_\sigma)\right|_{\eta_\sigma=\eta_{\sigma_s}}, \quad \omega(\eta_\sigma) = \partial f_{q_t}(\eta_\sigma)/\partial\eta_\sigma\Big|_{\eta_\sigma=\eta_{\sigma_s}}.$$

Here, τ_{cor} is the correlation time of a random field, which is a measure of the rate of random fluctuations of the medium and is defined as the memory time of the process:

$$\tau_{cor} = \int_0^\infty \frac{C(\tau)d\tau}{C_0}.$$

For the correlation function, $C(t) = C(0)\exp\{-\gamma_\tau t\}$, we obtain $\tau_{cor} = \gamma_\tau^{-1}$, where γ_τ is a constant. The fluctuating medium is characterized by significantly smaller correlation time τ_{cor} of a random process than the characteristic macroscopic time of the system τ_m [6]. In view of the technological conditions [1, 2] of obtaining glass-like semiconductors of the As-S(Se) system, $\tau_{cor} << \tau_m$ ($\tau_{cor} \approx 10^{-2}\ldots10^{-1}$ s, $\tau_m \approx 10^5$ s).

A distinctive feature of the white noise approach is that it is an entirely random process capable of acquiring an independent value at any time and having significant dispersion [12]. The energy is uniformly distributed over all frequencies of the white noise spectrum similar to the white light spectrum, and the correlation function is proportional to the Dirac δ-function [14]. The time evolution of the system transition probability density $\Re(Y, t+s|\eta_\sigma, s)$ during the periods s, $t = s$ is described by the Fokker–Planck equation (FPE) [12]. The Fokker–Planck equation in Stratonovich's interpretation looks as follows [12]:

$$\frac{d\Re(Y, t+s \mid \eta_\sigma, s)}{dt} = \\ = -\partial_Y\left[f_{q_t}(Y) + \frac{\sigma_\xi^{\ 2}}{2}g(Y)\partial_Y g(Y)\right]\Re(Y, t+s|\eta_\sigma, s) + \\ + \frac{\sigma_\xi^{\ 2}}{2}\partial_{YY} g^2 \Re(Y, t+s|\eta_\sigma, s). \tag{4}$$

Here, ∂_Y and ∂_{YY} are, respectively, the first and the second derivatives with respect to Y, and Y and η_σ are the system state parameters at the time points, respectively. The approach by Stratonovich enables direct modeling of the correlations between the random environment and the system [13].

2.2. Effect of white noise on the formation of self-organized structures

We examine a synergetic approach to the study of transition processes to non-crystalline state during cooling of non-crystalline system melts [8]. The following self-consistent system of equations for the fraction of atoms in the soft atomic configurations σ, the mean-square displacements of atoms $y_l(D_l^{\alpha\alpha})$ and $y_t(D_t^{\alpha\alpha})$, and the self-consistent interaction potentials $\tilde{f}, \tilde{g}$ was obtained in [16, 19]:

$$F_1(\sigma) = -\tilde{a}_0\tilde{q}\eta + c\eta^2 + b\eta^3, \; F_2(y_l) = \frac{\eta_{y_l}}{\tau_{y_l}}, \; F_3(y_t) = \frac{\eta_{y_t}}{\tau_{y_t}}. \tag{5}$$

Here, the functions $F_1(\sigma), F_2(y_l), F_3(y_t)$ are determined by the following equations [6, 19]:

$$F_1(\sigma) = \sigma\xi_P + (1-\sigma)z \times \\ \times\left\{\frac{e^{-y_l}}{2}\left[B(y_l) - \frac{P^* e^{y_l}}{12(1-\sigma)^2}\left(\frac{r}{a_0}\right)^2\right] + \frac{G_0/V_0}{(1+2y_t)}\right\} - \\ - z(1-\sigma)\sigma\left(1 + \frac{y_l}{4} - \frac{1}{6}\ln\frac{B(y_l)}{2}\right)A(y_l) - \tau\ln\frac{\frac{g_2}{\sigma}-1}{\frac{g_1}{1-\sigma}-1} = 0,$$

$$F_2(y_l) = \frac{3e^{-y_l}\tau}{8\sqrt{2}(1-\sigma)}\left[B(y_l) + \frac{P^* e^{y_l}}{6(1-\sigma)^2}\left(\frac{r}{a_0}\right)^2\right]^{-1} \times \\ \times\left(1 + \frac{0.022(1+2y_t)^{-2}}{e^{y_l}\left[B(y_l) + \frac{P^* e^{y_l}}{6(1-\sigma)^2}\left(\frac{r}{a_0}\right)^2\right]}\right) - y_l = 0,$$

$$F_3(y_t) = \frac{3(1+2y_t)\tau}{8(1-\sigma)e^{\frac{y_l}{2}}}\left[B(y_l) + \frac{P^* e^{y_l}}{6(1-\sigma)^2}\left(\frac{r}{a_0}\right)^2\right]^{-\frac{1}{2}} - y_t = 0.$$

Here, $\tau = \theta/V_0$ is the temperature and $\xi_P = \varphi/V_0$ is the anisotropy parameter of the soft and crystalline interaction potentials, respectively. The ratio $G_0/V_0 \approx 0.1$ is taken into account. The self-consistent system (5) enables studying dynamic stability and temperature behavior of systems with non-equilibrium phase transformations ($q \neq 0$) using the calculation results obtained for equilibrium transformations ($q = 0$) [19]. The values of the parameters $\sigma, y_l, y_t, \tilde{f}, \tilde{g}$ characterizing the system during the formation process change due to heat and mass transfer. Therefore, the latter characteristics also fluctuate and, most importantly, can be controlled [16]. Fluctuations in the medium, such as *e.g.* changes of the melt cooling rate q, influence the formation of self-organized structures and, more importantly, can generate qualitatively new non-equilibrium transitions.

We consider the influence of external white noise on the structure and dynamic stability of non-crystalline materials. Formation of a non-crystalline structure may be described by a bifurcation process characterized by the solutions of a system of nonlinear equations [8], where the external controlling parameter of the medium may be specified by the equation:

$$\lambda = \begin{cases} a \cdot \tilde{q} & \text{at}\, q \to q_c, \\ a \cdot \arctan(\ln[1+\tilde{q}]) & \text{at}\ q \neq q_c. \end{cases}$$

Here, q_c is the maximum cooling rate at which the non-crystalline phase is formed in the system and $\tilde{q} = \dfrac{q - q_c}{q_c}$ is the reduced cooling rate, respectively. We consider the case $\lambda < 0$ or $q > q_c$. SDE in Stratonovich's interpretation looks as follows:

$$d\eta_\sigma(t) = \left(\lambda\eta_\sigma + \gamma\eta_\sigma{}^2 - \beta\eta_\sigma{}^3\right)dt + \tilde{\sigma}_\xi \eta_\sigma dW_t. \tag{6}$$

Here, $dW_t = \xi_t dt$ is the increment of a random variable ξ_t, $\tilde{\sigma}_\xi = \sigma_\xi / q_c$ is the intensity of external noise reduced to the limit, and γ, β are the system parameters, respectively. The evolution of the distribution density function $\Re(\eta_\sigma, t)$ of the variable η_σ is presented as follows:

$$\partial_t \Re(\eta_\sigma, t) = -\partial_\eta \left(\lambda\eta_\sigma + \gamma\eta_\sigma^2 - \beta\eta_\sigma^3 + \frac{\tilde{\sigma}_\xi^2}{2}\eta_\sigma\right)\Re(\eta_\sigma, t) + \\ + \frac{\tilde{\sigma}_\xi^2}{2}\partial^2_{\eta\eta}\left(\eta_\sigma^2 \Re(\eta_\sigma, t)\right). \tag{7}$$

An external random field causes fluctuations of the parameters of the non-crystalline system. The internal fluctuations of the system parameters are accompanied by the fluctuations related to and caused by the fluctuations of the external temperature field. The indicated fluctuations of the system parameters are described by the distribution function $\Re(\eta_\sigma, t)$. The transition under the influence of external noise induces qualitative change of the system state function $\Re(\eta_\sigma, t)$. Only transitions caused by external noise are considered. In this case, the parameters defining the transition include the mean value of the external white noise intensity, variance, and correlation time.

The continuity equation takes the following form:

$$\partial_t \Re(\eta_\sigma, t) + \partial_{\eta_\sigma} J(\eta_\sigma, t) = 0, \tag{8}$$

where

$$J(\eta_\sigma, t) = \left(\lambda\eta_\sigma + \gamma\eta_\sigma^2 - \beta\eta_\sigma^3 + \frac{\tilde{\sigma}_\xi^2}{2}\eta_\sigma\right)\Re(\eta_\sigma, t) - \\ - \frac{\tilde{\sigma}_\xi^2}{2}\partial_{\eta_\sigma}\left(\eta_\sigma^2 \Re(\eta_\sigma, t)\right)$$

is the flux of the soft atomic configurations of the non-crystalline system [19].

In the stationary case, $\partial_{\eta_\sigma} J(\eta_\sigma) = 0$ and $\partial_t \Re(\eta_\sigma) = 0$. The stationary density of the probability distribution function $\Re_s(\eta_\sigma)$ corresponding to the stationary behavior of the system is determined from the following relation:

$$\Re_s(\eta_\sigma) = \frac{N_{norm}}{g(\eta_\sigma)} \exp\left\{\frac{2\lambda}{\tilde{\sigma}_\xi^2} - \frac{\lambda(u) + h(u)}{g(u)}\right\} = \\ = N_{norm}\eta_\sigma^{\frac{2\lambda}{\tilde{\sigma}_\xi^2} - 1} \exp\left\{\frac{2\gamma}{\tilde{\sigma}_\xi^2}\eta_\sigma - \frac{\beta}{\tilde{\sigma}_\xi^2}\eta_\sigma^2\right\}. \tag{9}$$

Here, N_{norm} is a normalizing coefficient calculated from the following relation:

$$\int_0^\infty \Re_s(\eta_\sigma) d\eta_\sigma = 1,$$

$$1/N_{norm} = \frac{1}{2}\left(\frac{\tilde{\sigma}_\xi^2}{\beta}\right)^{\frac{\lambda}{\sigma^2}} \Gamma\left(\frac{\lambda}{\tilde{\sigma}_\xi^2}\right) + \frac{\gamma}{\tilde{\sigma}_\xi^2}\left(\frac{\tilde{\sigma}_\xi^2}{\beta}\right)^{\frac{\lambda}{\tilde{\sigma}_\xi^2} + \frac{1}{2}} \Gamma\left(\frac{\lambda}{\tilde{\sigma}_\xi^2} + \frac{1}{2}\right), \tag{10}$$

where $\Gamma(x)$ is the Gamma-function. It follows from (9) and (10) that the function $\Re_s(\eta_\sigma)$ is integrated over the interval (0, 1). That is, stationary solutions for the non-crystalline state exist only at $\dfrac{2\lambda}{\tilde{\sigma}_\xi^2 - 1} > 1$ or $\lambda > 0$ ($q > q_c$). At $\lambda < 0$ or, identically, $\tilde{q} < 0$ and $q < q_c$, the probability density (10) behaves as the δ-function, as follows from the expression for N_{norm}.

Extremums of the stationary density $\Re_s(\eta_\sigma)$ correspond to the macroscopic stationary non-equilibrium states of the system. Qualitative change of the form of $\Re_s(\eta_\sigma)$ with the change of the control parameter and the intensity of noise indicates the transition. In fact, we rewrite (2) in terms of the synergetic potential $V_{q_t}(\eta_\sigma)$ of soft atomic configurations $V_{q_t}(\eta_\sigma) = -\int_0^{\eta_\sigma}(h(z)+q_t g(z))dz$ as follows:

$$\frac{\partial\eta_\sigma}{\partial t} = -\frac{\partial}{\partial\eta_\sigma}V_{q_t}(\eta_\sigma). \tag{11}$$

Eq. (11) can be also derived from the Landau–Halatnikov equation [12, 19]. Stationary states of the system are defined by zero values of the right-hand side of Eq. (11) and the corresponding potential $V_{q_t}(\eta_\sigma)$. Steady modes $\omega(\eta_\sigma)$ of the system behavior in the linear approximation of the stability theory [13] are defined through the potential $V_{q_t}(\eta_\sigma)$ as $\omega(\eta_\sigma) = -\frac{\partial^2}{\partial\eta_\sigma^{\ 2}}V_{q_t}(\eta_\sigma)$.

We consider the way the external white noise effects the behavior of the synergetic potential $V_{q_t}(\eta_\sigma)$ in the deterministic and stochastic cases. In the deterministic case, the stationary probability density $\Re_s(\eta_\sigma)$ consists of the δ-speed peaks centered near stationary states. External noise causes expansion of the δ-like peaks. External fluctuations induce minor fluctuations of the soft atomic configurations in the vicinity of the positions of the stationary states and cause percolation of the distribution of the soft atomic configurations. The percolation increases the diffusion component ($\approx \tilde{\sigma}_\xi^2 \cdot g(\eta_\sigma)$, see Eq. (7)) and facilitates rearrangement of the soft atomic configurations at the middle order level. Therefore, a decrease in orderliness at the level of short-range order and an increase in orderliness at the nanolevel of medium order are observed. Such phenomena are related to fractality through the influence of external noise manifested as the synchronicity and coherence of the transformation of ordering at different spatial scales. Hence, rearrangement does not occur separately at each spatial level, but rather fractally. It is also one of the unique opportunities for the development of intelligent semiconductor materials based on non-crystalline systems [18, 19]). The equation for determining the extremes of the stationary probability density (9) at $q \to q_c$ looks as follows:

$$\lambda\eta_{\sigma m} + \gamma\eta_{\sigma m}^2 - \beta\eta_{\sigma m}^3 - \frac{\tilde{\sigma}_\xi^2}{2}\eta_{\sigma m} = 0$$

and its solutions are

$$\eta_{\sigma 1} = 0,\ \eta_{\sigma 2,3} = \frac{\gamma \pm \sqrt{\gamma^2 + 4\beta\left(\lambda - \tilde{\sigma}_\xi^2/2\right)}}{2\beta}. \tag{12}$$

The solutions $\eta_{\sigma 2,3}$ exist at $\lambda > \frac{\tilde{\sigma}_\xi^2}{2} - \frac{\gamma^2}{4\beta}$ and always correspond to the maximum of $\Re_s(\eta_\sigma)$. The solution $\eta_{\sigma 1}$ corresponds to the maximum only at $0 < \lambda < \tilde{\sigma}_\xi^2/2$ (see Fig. 1).

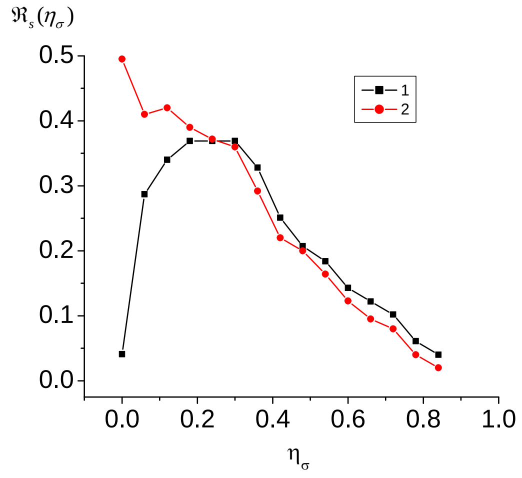

Fig. 1. The dependence of the stationary density on the distribution of the relative change in the temperature in the loss of dynamic stability $\Re_S(\eta_\sigma)$. $\sigma_\xi = 1.5 \cdot 10^{-2}$ K/s, $\tilde{q} = 0.7$ (red circle), $\tilde{q} = 1.0$ (black square).

Hence, qualitatively new transitions to the non-crystalline state can be realized in the presence of a random temperature field. The transition at $\sigma_\xi = 0$ and $\lambda = 0$ corresponds to the deterministic case $(q \le q_c)$. The transition at $\lambda \ne 0$ and $\tilde{\sigma}_\xi > \tilde{\sigma}_{\xi c} = (2\lambda)^{1/2}$ is accompanied by a sharp change of the form of the probability density with a blurring δ-like distribution $\Re_s(\eta_\sigma)$ in the direction of non-zero values of η_σ (see Fig. 1).

3. White noise: Bifurcation diagram

3.1. Bifurcation diagram

Behavior of the stationary density of the distribution function of the relative temperature change under the condition of the loss of dynamic stability as a function of external noise intensity is shown in Fig. 2. The probability density has the following features. If the velocity q is less than the critical one ($\lambda < 0$), the stationary point $\eta_\sigma = 0$ corresponding to the equilibrium crystalline state is an asymptotic constant (the distribution $\Re_s(\eta_\sigma)$ in this case behaves like the δ-function, see Fig. 2).

The stationary probability density goes to infinity at $\eta_\sigma \to 0$ in the presence of external noise and at the values of the control parameter $0 < \lambda < \tilde{\sigma}_\xi/2$. That is, the reproducible part of the δ-function of the properties and $\eta_\sigma = 0$ is the most probable value. However, $\Re_s(\eta_\sigma) \ne 0$ at $\eta_\sigma \ne 0$ (see Fig. 2). In such a case, transition to a partially disordered state is realized at $t \to \infty$ with possibility of relaxation to equilibrium. Consequently, the transition to a partially disordered state is realized at $t \to \infty$ in this case corresponding to the relaxation to equilibrium. A non-equilibrium structure

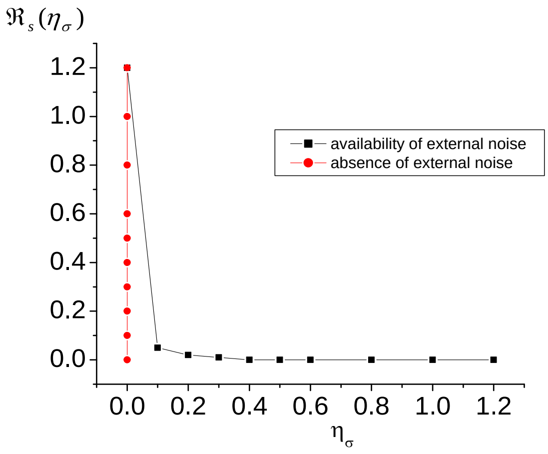

Fig. 2. Qualitative change in the dependence nature $\Re_s(\eta_\sigma)$ for $\tilde{q} = 0$ in the absence (red circle) and presence (black square) of the external noise. $\sigma_\xi = 0.5 \cdot 10^{-2}$ K/s.

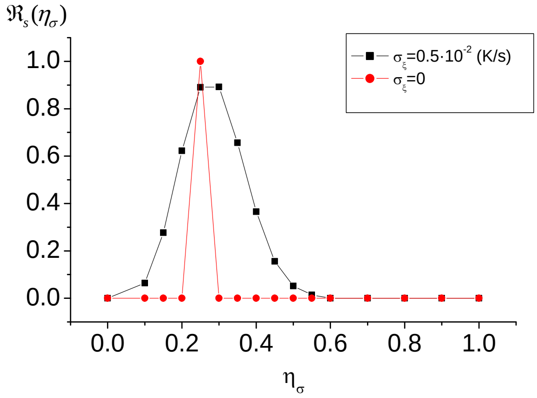

Fig. 3. Changing the nature of the distribution function dependence $\Re_s(\eta_\sigma, z)$ in the absence (red circle) and the presence of external noise (black square) for $\tilde{q} \geq \tilde{q}_c$ ($\tilde{q} = 1$, $\sigma_\xi = 0.5 \cdot 10^{-2}$ K/s).

may also form since $\Re_s(\eta_\sigma)$ has a local maximum at $\eta_\sigma \neq 0$ but is not asymptotically stable (see Fig. 2). This structure is a qualitatively new state of the crystalline system, which is caused by external noise. In this case, an accidental temperature field has a partially disruptive effect during obtaining the system.

At $\lambda = \tilde{\sigma}_\xi^2 / 2$, the distribution pattern $\Re_s(\eta_\sigma)$ radically changes again. Although the probability density is different from zero at η_σ, the most probable value of $\Re_s(\eta_\sigma)$ is achieved at $\eta_\sigma \neq 0$ (see Fig. 3). At $\lambda \neq 0$ and $\tilde{\sigma}_\xi > \tilde{\sigma}_{\xi c} = (2\lambda)^{1/2}$, the formation probability of an equilibrium amorphous-crystalline structure tends to zero. $\Re_s(\eta_\sigma)$ has a distinct nonzero extreme value at this. Therefore, the transition to the non-crystalline state in the presence of an external random temperature field shows qualitative changes in the behavior of the system when the noise intensity changes as compared to the deterministic case. This testifies presence of two types of noise-induced transitions in the system under consideration, which are absent under deterministic actions, namely the shift of the deterministic bifurcation diagram and the formation of new levels of system structuring. Qualitative change in the behavior of the system, which is described by the distribution function $\Re(\eta_\sigma, t)$, is observed at the intensities greater than the threshold values $\sigma_\xi \geq \sigma_{\xi c}$ (in the deterministic case with $q \geq q_c$).

The effect of environmental fluctuations depends on the state of the system ($g(\eta_\sigma) \neq 1$) (see Fig. 3). It means that not only disordering at the short-range order level, which is manifested as the change of atom fractions in the soft atomic configurations near the mean value, is present but also rearrangement at the medium order level resulted from the change of the extremum position of $\Re(\eta_\sigma, z)$. With the increase in noise intensity at $\sigma_\xi \geq \sigma_{\xi c}$, the summand $\approx g(\eta_\sigma)$ in $V_{q_t}(\eta_\sigma)$ (see Eq. (11)) is no more small. It causes diffusional redistribution of the soft atomic configuration areas and, hence, a qualitative change in the system state. Therefore, multiplicative white noise can induce formation of qualitatively new self-organized structures. The system no longer adapts its behavior to the average properties of the environment and the effects of external noise at $q_t = q + \sigma_\xi \xi_t$ and $\bar{q}_t$ are not equivalent. This means that the average value of the cooling rate alone is not sufficient to predict the properties of the system. At the external noise intensities $\sigma_\xi << \sigma_{\xi c}$, the situation is similar to the deterministic case. The system behavior is characterized by the mean values of the environmental parameters. In this case, the effects of external noise on the system at $q_t = q + \sigma_\xi \xi_t$ and $q_t = \bar{q}_t$ are qualitatively the same. The results of the study of the influence of the diffusion component of external noise on the behavior of the distribution function are shown in Fig. 4. It is obvious

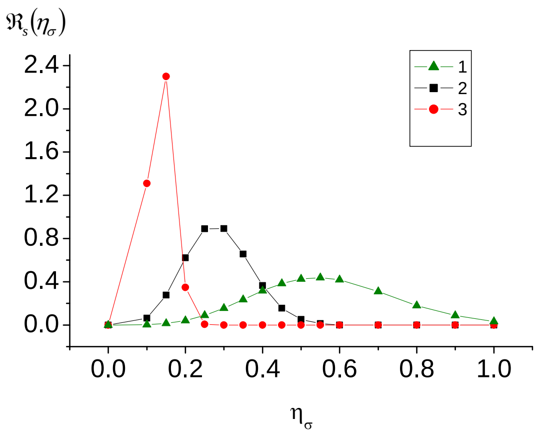

Fig. 4. Influence of the parameter γ diffusion component on the nature of the distribution function $\Re_s(\eta_\sigma)$. $\tilde{q} = 1$, $\alpha/\beta = 0.1$, *1* – $\gamma/\beta = 0.5$, *2* – 0.1, *3* – 0.01 for $\sigma_\xi = 0.5 \cdot 10^{-2}$ K/s.

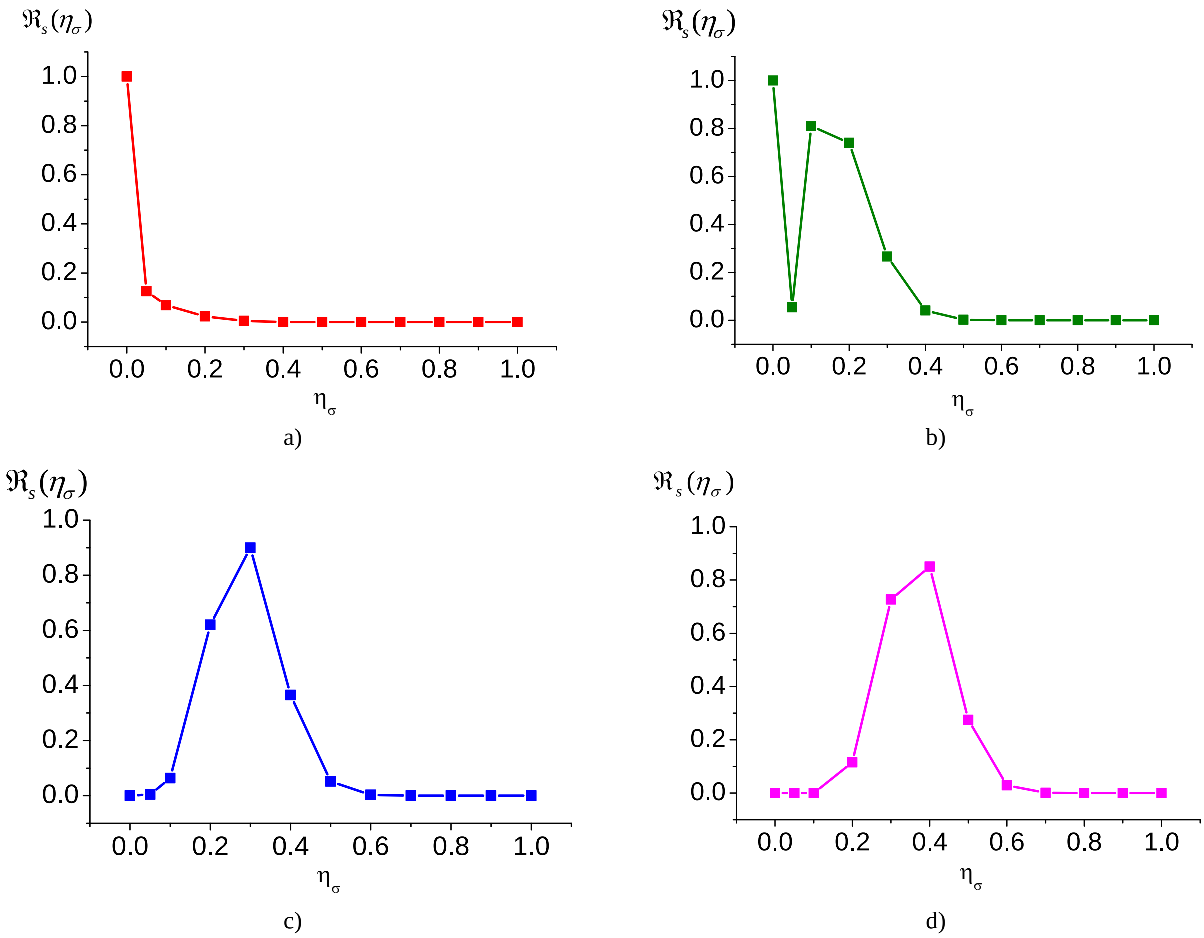

Fig. 5. Dependence of the stationary density of the distribution function $\Re_s(\eta_\sigma)$ at different cooling velocity $\tilde{q}$ and noise intensity $\sigma_\xi = 0.5\cdot10^{-2}$ K/s. a) $\tilde{q} = 0$, b) 0.2, c)1, d) 5.

from this figure that increase of γ/β at constant $\tilde{q},\tilde{\sigma}_\xi$ leads to shift of the extreme value of the distribution function towards higher values of η_σ and spillage of $\Re_s$ around the extreme value.

Figs 5 and 6 present the dependence $\Re_s(\eta_\sigma)$ at various reduced cooling rates $\tilde{q}$ and external noise intensities σ_ξ. Qualitative state of the system is determined by the number and positions of the extremums of the probability density $\Re_s(\eta_\sigma)$ in the stochastic case and synergetic potential $V_{q_t}(\eta_\sigma)$ in the deterministic case. The extremums of $\Re_s(\eta_\sigma)$ correspond to the system states (attractors). One maximum of the stationary density (see Figs 5 and 6) indicates that the system fluctuates relative to one macroscopic state (attractor). If the stationary density has two or more maxima at the same values of external noise, the system fluctuates between two possible states.

We analyze the dependence of the stationary density extremums on the external parameter values at different noise intensities (see Figs 5 and 6).

The dependence of the extremums of $\Re_s(\eta_\sigma)$ on $\tilde{q}$ can be regarded as a modification of the deterministic bifurcation diagram with a shift of the curve $\eta_\sigma(\tilde{q})$ caused by $\tilde{q}$ and $\tilde{\sigma}_\xi^2/2$. At the bifurcation point, the system defines a self-organized self-sufficient structure and the corresponding type of self-organization, while after the bifurcation, the system obeys macroscopic kinetic laws. In the vicinity of the bifurcation, different ways of system evolution are equally probable, leading to coexistence of different types of ordering. The system forms a certain non-crystalline structure upon making a random synergetic choice (see Fig. 7). The stochastic variability of the fabrication conditions in the medium leads to percolation of the system states to a certain average value. The notion that the role of fluctuations is ultimately secondary is based on the linear relationship between the system and the environment. However, the theoretical studies of melt cooling in a fluctuating medium with noise presented above indicate just the opposite, namely that the random nature of the medium

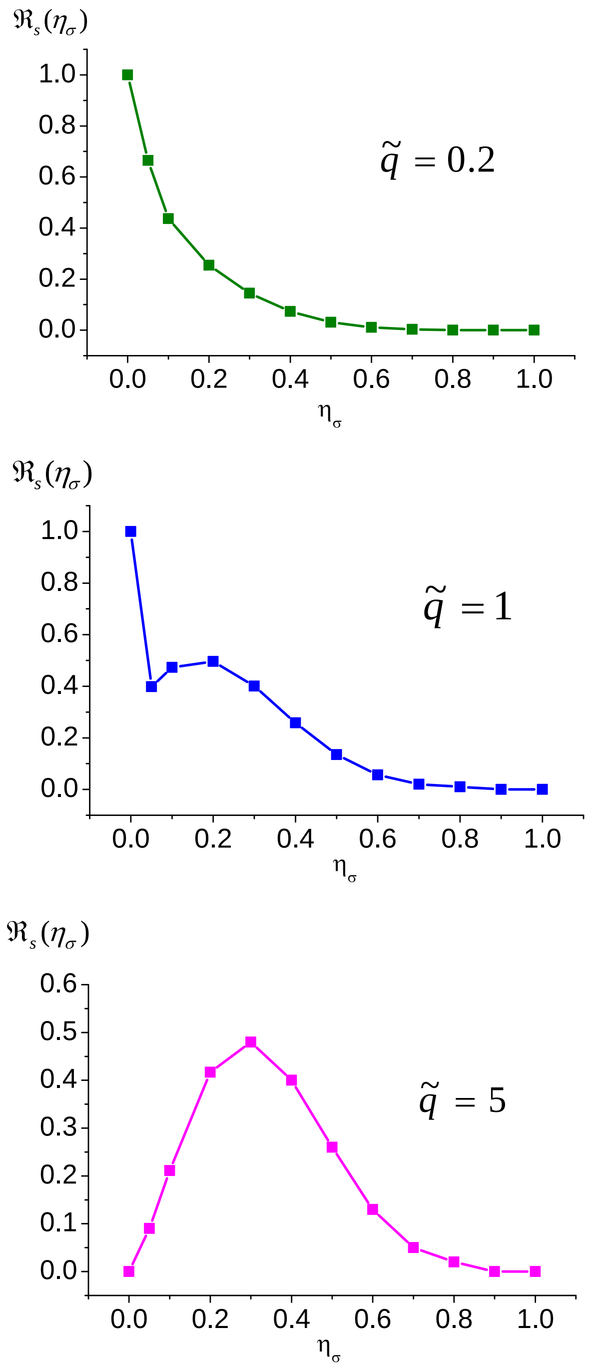

Fig. 6. Dependence of the stationary density of the distribution function $\Re_s(\eta_\sigma)$ at different cooling velocity $\tilde{q}$ and noise intensity $\sigma_\xi = 1\cdot10^{-2}$ K/s.

along with the disorganizing effect can initiate formation of qualitatively new self-organized structures of non-crystalline materials. The spectrum of structures obtained in this way is significantly larger than the one possible to obtain under corresponding deterministic conditions (see Figs 5 and 6). Therefore, enhancement of the stochastic variability of the medium enables controlling the fractal structure of a non-crystalline system depending on the noise intensity.

3.2. Comparison of model behavior with experimental data

The presented distributions $\Re_s(\eta_\sigma)$ (see Figs 5 and 6) show that transition to the non-crystalline state can be achieved by maintaining the cooling rate constant and increasing the intensity of fluctuations in the external temperature field. The distribution function has an extremum at fixed values $\lambda \neq 0$ and intensities of noise above the threshold value $\tilde{\sigma}_\xi > \tilde{\sigma}_{\xi c} = (2\lambda)^{1/2}$. We obtain $\Re_s(0) = 0$. Hence, there is a clearly defined non-zero value $\eta_\sigma \neq 0$ at which the distribution function reaches an extremum. At $\lambda = \tilde{\sigma}^2{}_\xi/2$, the function $\Re_s(0)$ remains finite and sharply increases ($\Re_s(0) \to \infty$) at the noise intensity $\lambda < \tilde{\sigma}^2{}_\xi/2$. Hence, no transition to a non-crystalline state with the formation of self-organized structures occurs. The peculiarity of noise-induced transitions as compared to the deterministic case is the possibility of realizing a set of values η_σ and, consequently, obtaining non-crystalline structures with different additional types of ordering [16].

The suggested approach allows calculating the critical values of parameter fluctuations when the structural characteristics of the non-crystalline materials are not sensitive to the changes of the conditions of their obtaining. Furthermore, this approach enables predicting the formation of qualitatively new self-organized structures at certain noise intensities. We consider the dependence of the threshold noise intensity on the cooling rate for non-crystalline As_2S_3 (critical cooling rate equals as $q_c = 5\cdot10^{-3}$ K/s [1, 4]). Therefore, the threshold noise intensity is $\sigma_{\xi c} = 3\cdot10^{-3}$ K/s at the cooling rate $q = 1.5\cdot10^{-2}$ K/s, $\tilde{q} = \dfrac{q-q_c}{q_c} = 2$. The system state changes during transition to the non-crystalline state in

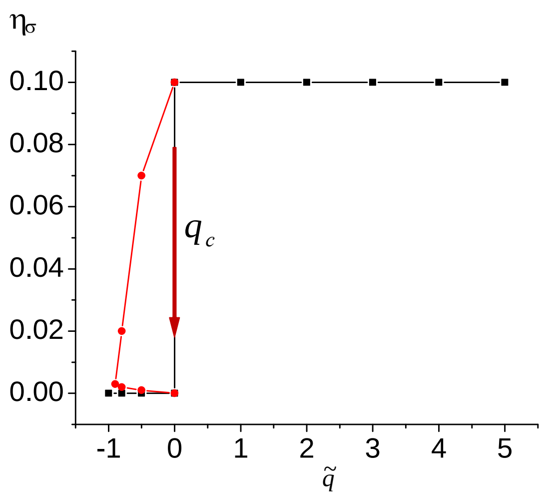

Fig. 7. Dependence of extremums of the distribution function $\Re_s(\eta_\sigma)$ on the cooling velocity q_c. $\sigma_\xi = 6\cdot10^{-3}$ K/s.

Table. Experimental and theoretical data on the influence of random temperature field on the stability of non-crystalline solid As_2S_3: $q = 1.5\cdot10^{-2}$ K/s, $q_c = 5\cdot10^{-3}$ K/s, $\tilde{q} = 2$, $T_g = 447$ K, and $\Delta T_g = \frac{T_g - T_{g\xi}}{T_g}$.

Intensity of noise	Experiment			Calculation
	T_g, K	ρ, g/cm³	ΔT_g, relative units	ΔT_g, relative units
$\sigma_\xi = 0$	447	3.192	0	0
$\sigma_\xi = 4\cdot10^{-3}$ K/s	445	3.190	0.005	0.009
$\sigma_\xi = 8\cdot10^{-3}$ K/s	442	3.185	0.011	0.015

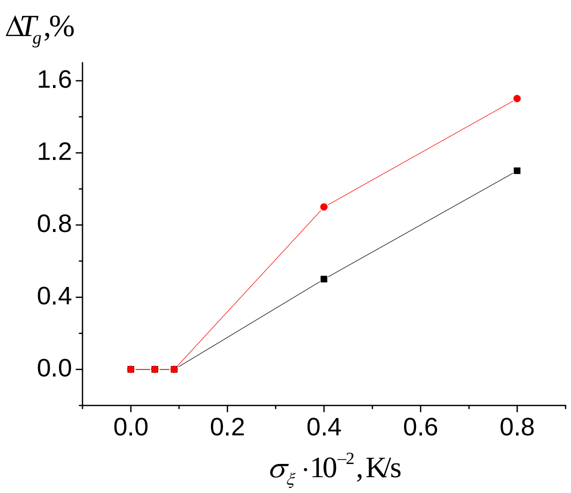

Fig. 8. Bifurcation diagram in plane $\{\Delta T_g, \sigma_\xi\}$ for non-crystalline As_2S_3 (red circle – theory, black square – experiment).

the deterministic case (fluctuations of the controlling medium parameter $\sigma_\xi < \sigma_{\xi c}$). This change is caused exclusively by the external parameter, namely the cooling rate q. For noise intensities $\sigma_\xi > \sigma_{\xi c}$, two parameters, namely the noise intensity σ_ξ and the cooling rate q must be taken into account. According to the theory of bifurcations [12, 19], this is manifested by the change of the attractor type, to which the system tends in the equilibrium ($q < q_c$) and non-equilibrium state ($q \geq q_c$, $\sigma_\xi > \sigma_{\xi c}$), and is described by the bifurcation diagram.

We analyze the effect of a random temperature field during cooling on structurally sensitive parameters of As_2S_3, namely its softening temperature T_g and density ρ. The random temperature field was set by changing the airflow intensity during cooling (see Table). As can be seen from this table, non-crystalline As_2S_3 materials obtained in the presence of an external random temperature field are characterized by the modified structurally sensitive parameters. Moreover, the change of the softening temperature of non-crystalline As_2S_3 under the influence of white noise has a bifurcation character on the plane $\{\Delta T_g, \sigma_\xi\}$ (see Fig. 8).

The research results described in this article enable improvement the understanding of a number of physical effects and processes in non-crystalline solids under the influence of external factors. Practical application aspect of our research is related to the study of the processes of self-organization and functional ordering in glassy materials aimed at predicting specified properties, providing the elemental base for solid and functional electronics, and developing smart materials for artificial intelligence based on semiconductor compounds [18, 20]. The fractality effect of external noise provides a unique practical insight [18, 21–24]. Moreover, synergy and object-oriented modeling [10, 21] enable the most adequate reproduction of the features and characteristics of the influence of external noise on the formation of non-crystalline systems.

4. Conclusions

• Study of the impact of the fluctuations of cooling rate on the formation of self-organized structures of non-crystalline materials in the white noise approximation enables determining the threshold values of noise intensity $\sigma_\xi < \sigma_{\xi c}$, at which the structural characteristics of non-crystalline materials are not sensitive to the changes in the conditions of their fabrication. It also enables predicting the formation of self-organized structures at the noise intensities $\sigma_\xi \geq \sigma_{\xi c}$.

• It has been found out from the analysis of the distribution function of the relative temperature change under the condition of the loss of dynamic stability that transition to the non-crystalline state may be achieved by maintaining a constant cooling rate but increasing the intensity of fluctuations of the external temperature field above the threshold value $\sigma_\xi \geq \sigma_{\xi c}$.

• We also found the presence of a noise-induced transition associated with the shift of the deterministic bifurcation diagram. The qualitative change in the system behavior described by the distribution function $\Re(\eta_\sigma)$ is observed at the intensities exceeding the threshold value. The state of the system is determined by the number and positions of extremums of the probability density $\Re(\eta_\sigma)$.

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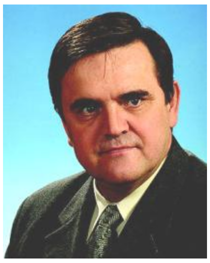

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Authors' contributions

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NataliyaYurkovych: conceptualization, methodology, validation, formal analysis, investigation, resources, data curation, writing – vizualization, review & editing, project administration.

Vladimir Seben: investigation, resources, writing – review & editing.

Самоорганізовані структури та нанорозмірні рівні їх формування в некристалічних напівпровідниках системи As-S(Se), індуковані білим шумом

М.І. Мар'ян, Н.В. Юркович, В. Шебень

Анотація. Розглянуто сингулярність та самоорганізуючий ефект нестабільності і випадковості при впливі зовнішнього білого шуму на утворення некристалічних матеріалів. Встановлено, що випадковий характер середовища одержання разом з дезорганізуючим ефектом може ініціювати утворення якісно нових самоорганізованих структур у некристалічних твердих тілах. Дослідження впливу випадкового температурного поля, прикладеного до розплаву в процесі охолодження, проведено для некристалічних напівпровідників системAs-S(Se). Знайдено умови, за яких некристалічна система у флуктуаційному зовнішньому середовищі підпорядковує свої властивості середнім властивостям середовища і відповідає детермінованому випадку. Також визначено умови, коли система реагує на випадкове середовище одержання не адитивно, а, навпаки, формує новий режим перетворення енергії самоорганізованої структури на нанорозмірному рівні. Отриманий таким чином спектр структур якісно більш різноманітний порівняно з можливим спектром за відповідних детермінованих умов.

Ключові слова: біфуркаційна діаграма, фрактальність, нанорозмірні ефекти, синергетика, білий шум, некристалічні напівпровідники, самоорганізовані структури.