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Formation and Modeling of Nanosized Levels of the Self-organized Structures in the Non-crystalline Thin Films of Ge-As-Te(S, Se) Systems

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A model of thermal instability and the formation of self-organized structures under the action of continuous infrared radiation of a CO_2-laser on amorphous condensates of Ge-As-Te (S, Se) systems applied to quartz or mica substrates is proposed, and its analysis is based on experimental studies. It is shown that the uniform distribution of the temperature field along the beam section at the power density of the radiation $P > P_c$ ($P_c = 1.8$-8.5 W/cm^2) becomes unstable and a radial-ring structure with a spatially inhomogeneous profile of the temperature field is formed. It is established that the bifurcation of thermal instability and the formation of a non-uniform temperature profile are carried out at the expense of self-regulating mechanisms, which lead to saturation of the absorption nonlinearity and equalization of the growth rate of the absorbed energy by heat transfer into the amorphous layer. Dependence of the threshold density of the infrared radiation power of the formation of ordered radial-ring structures on the irradiation duration is investigated. A bifurcation diagram in a plane *{radiation power (P), temperature (T)}* that describes the singular behavior of optical density depending on the duration of exposure to radiation is constructed. It is shown that a radial-ring structure with the number of rays $m \geq 3$ is formed for amorphous condensates of Ge-As-Te (S, Se) systems at irradiation power densities $P \geq 18.6$ W/cm^2, which is consistent with the experimental data. The peculiarities of the behavior of self-organized structures in thin-film layers are analyzed, namely the sensitivity of the system to the technological conditions of obtaining, the parameters of electromagnetic radiation and the possibility of realization of the butterfly effect. The ways of formation and implementation of fractality by nanosized levels of structuring and lifetime for the self-organized structures in the presented non-crystalline materials of Ge-As-Te (S, Se) systems are considered.

Keywords: Effect of infrared irradiation, Fractality, Non-crystalline thin films, Self-organized structures, Synergetics.

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1. INTRODUCTION

Amorphous thin-film chalcogenides and their multicomponent alloys are very interesting objects with wide practical and fundamental applications [1-5]. Due to the optical transparency in the near, medium and far infrared regions, their significant nonlinearity, chalcogenide non-crystalline materials are used as active and passive elements in optics and sensory devices [2-4], telecommunications as a thermal image and the generation of nonlinear light [1], cyber security systems [6]. Intensive and reverse crystallization in thin-film systems of chalcogenides is the basis for the use of these materials in the creation of non-volatile memory [4]. The mentioned applications of the chalcogenide materials of Ge(As)-Te (S, Se) systems could not be realized without the knowledge of the basic properties and processes occurring in these surprising materials [6-8]. In addition, the fundamental researches of the self-organizing processes and formation of self-organized structures in them, and into the influence of infrared irradiation using synergetics are extremely important and unique [4, 9-11]. One example of the systems for which significant deviations from the equilibrium state are accompanied by the formation of self-organized structures is the realization of space-time structures as a result of thermal instabilities under the action of ultra-short pulses of nano- and picosecond duration, or continuous infrared (IR) radiation in semiconductor layers [4, 12]. A special feature of these structures is the sensitivity of the system to the technological conditions of obtaining, parameters of electromagnetic radiation and the possibility of realizing the butterfly effect [6, 13]. From this perspective, the study of the processes of the formation of ordered self-organized structures in thin-film systems continues to arouse lasting and constant interest in the synergetic approach and its fundamental application on problems of energy conservation, fractality of the nanosized levels of structuring [13-15].

The paper presents effect of continuous IR radiation of a CO_2-laser on thin-film semiconductor structures of systems Ge-As-Te (S, Se) with the use of synergetic approach. The aim of the research is to establish the possibility of forming the self-organized structures and the corresponding implementation of anticipated their structural-sensitive properties.

2. THERMAL INSTABILITIES IN THE NON-CRYSTALLINE MATERIALS: THE EFFECT OF IR IRRADIATION

2.1 The Concepts and Investigation Methods

We consider a system that contains a substrate and a layer of the non-crystalline material of Ge-As-Te (S, Se) systems applied to it. Peculiarities of the behavior of the properties of telurofast transparent materials in the IR region are determined by absorption in present the region of the wavelengths $\Lambda = 1060$ nm of the

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substrate on which they are applied, and the mechanism of heat dissipation. Radiation with power density

$$P(r) = I(r)/s_0$$

with the Gaussian distribution along the beam section

$$I(r) = I_0 \,\boldsymbol{exp}\left\{-r^2/r_0^2\right\}$$

(r_0 is an effective radius, $s_0 = \pi r_0$ is an effective area of the beam cross section) and t_p is the duration of the exposure, falls normally to the surface of the layer and is completely absorbed along the z axis in the volume of the substrate. In the IR region of the spectrum, a significant temperature dependence of the absorption cross-section of the substrate radiation is observed [1, 16].

The absorption capacity of the substrates can be approximated by expression

$$\gamma = \gamma_0 \,\boldsymbol{exp}\left\{-\alpha_a \cdot T_a/T\right\},$$

where γ_0 is the absorption coefficient before irradiation, α_α and T_a are the parameters of the absorption capacity of the substrate, T is the temperature. Self-consistent increase in absorption capacity and the establishment of a non-stationary temperature regime on the boundary with an amorphous layer can lead to the emergence and development of thermal instability [14, 15]. In order to study the formation of self-organized structures due to thermal instability, let us consider the dynamics of temperature change of the "substrate-layer" system, namely the temperature distribution on the surface with the amorphous layer and in the substrate. This problem is described by a system of nonlinear differential equations that takes into account the processes of heat generation, its propagation and heat transfer for the substrate and the thin-film amorphous layer:

$$\rho C\frac{\partial T}{\partial t} = div\left(\chi \cdot \boldsymbol{grad(T)}\right) + W(T) - Q(T), \qquad (1)$$

$$\rho_a C_a \frac{\partial T}{\partial t} = div\left(\chi_a \cdot \boldsymbol{grad(T)}\right) + Q(T) - Q_a\boldsymbol{(T)}.$$

Here C, ρ and χ are the heat capacity, density and thermal conductivity of the substrate respectively; $W(T) = \gamma UP\,\boldsymbol{exp}(-\gamma \cdot z)f(t)$ is the laser heat source; U is the optical transmission of the amorphous layer at the wavelength $\Lambda = 1060$ nm; $f(t) = 1$ at $t < t_p$ and $f(t) = 0$ at $t > t_p$;

$$Q(T) = \int_{T_0}^{T} \eta_h\boldsymbol{(T)}\frac{s_0}{V}dT = \eta_h\boldsymbol{(}T - T_0\boldsymbol{)}/d_\gamma$$

is the heat exchange with amorphous layer (η_h is the heat transfer constant, T_0 is the temperature before irradiation, $V = s_0 d_\gamma$ is the effective volume of the absorption radiation, d_γ is the effective depth of radiation penetration into the substrate); C_α, ρ_α and χ_a are the heat capacity, density and thermal conductivity of the amorphous layer, respectively; $Q_a\boldsymbol{(T)} = L_a\rho_a f_a$ is the quantity of heat that is absorbed during the crystallization of the amorphous layer, L_a is the heat of crystallization, $f_a = \upsilon\,\boldsymbol{exp(}-\upsilon \cdot t_p\boldsymbol{)}$ is the proportion of crystallized volume per unit of time, which is determined by the Johnson-Mel-Abraham equation

$$\upsilon = \upsilon_0 \,\boldsymbol{exp(}{}^{E_a}\!\big/\!{}_{k_B T}\boldsymbol{)},$$

E_a is the activation energy of crystallization [16].

Initial and boundary conditions are the following:

$$\left.\frac{\partial T(r,z,t)}{\partial z}\right|_{z\to\infty} = 0,$$

$$T(r,z,t)\big|_{r\to\pm\infty} = T_0,\ T(r,z,t)\big|_{t=0} = T_0,$$

$$\chi \cdot \left.\frac{\partial T(r,z,t)}{\partial z}\right|_{z=0} = \eta_h \cdot \left(T\boldsymbol{(}r,z,t\boldsymbol{)} - T_{substrate}\boldsymbol{(}r,z,t\boldsymbol{)}\right)\Big|_{z=0}.$$

The depth of radiation penetration d_γ into the quartz or mica substrate at the wavelength $\Lambda = 1060$ nm is about $d_\gamma \leq 100$ nm, while the thermal front extends over the entire depth of the substrate $d \approx 10^3$ μm >> d_γ and through heat transfer to the amorphous layer. As γ increases sharply with temperature, one can use the approximation

$$\boldsymbol{exp(}-\gamma z\boldsymbol{)} \approx \delta(z) = \begin{cases} 1, & z \leq d_\gamma, \\ 0, & z > d_\gamma, \end{cases}$$

that is the radiation is completely absorbed in the near-surface layer of the substrate d_γ.

To solve the equation (1) and to analyze the results obtained, it is convenient to apply dimensionless quantities:

$$\Phi = \frac{T}{T_a},\ \Phi_0 = \frac{T_0}{T_a},\ \tilde{p} = \frac{UP}{\eta_h T_a},\ \tau_t = \frac{\eta_h \cdot t}{\rho C d_\gamma},\ \tilde{k} = \frac{\chi \cdot d_\gamma}{\eta_h r_0^2}.$$

The equation (1) for the temperature distribution in the substrate takes the form:

$$\frac{\partial\Phi}{\partial\tau_t} = \tilde{k}\nabla^2\Phi + \tilde{p}\,\boldsymbol{exp}\left\{-\frac{\alpha_a}{\Phi}\right\}\boldsymbol{exp(}-\frac{r^2}{r_0^2}\boldsymbol{)}\delta\boldsymbol{(}z\boldsymbol{)}f(\tau_t) - \Phi + \Phi_0. \qquad (2)$$

Here ∇^2 is the Laplace's operator in a cylindrical coordinate system (r, φ, z). The stationary temperature field Φ_s, which determines the bifurcation diagram $\left\{\tilde{p},\Phi\right\}$ on the surface of the substrate and the amorphous layer $z = 0$, is given by the equation

$$\tilde{k}\nabla^2\Phi = -\tilde{p}\,\boldsymbol{exp}\left\{-\frac{\alpha_a}{\Phi}\right\}\boldsymbol{exp(}-\frac{r^2}{r_0^2}\boldsymbol{)} + \Phi - \Phi_0. \qquad (3)$$

The configuration of the stationary temperature field depends on the geometry of the two-layer system "amorphous layer + substrate", the density of the heat source and heat removal. The system has instability by the parameter $\tilde{p}$ associated with the power density of the radiation P. Depending on $\tilde{p}$, there are either one or three stationary states that differ in the driven temperature Φ. The type of singular points on a bifurcation diagram $\{\tilde{p}, \Phi\}$ and their stability are investigated by the expansion of small deviations Φ from stationary solutions $\Phi = \Phi_s + \delta\Phi$ where the solution of equation (3) is Φ_s, the variation of function Φ is $\delta\Phi$ [17, 18]. We substitute $\Phi = \Phi_s + \delta\Phi$ into equation (2) and keep only linear in $\delta\Phi$ terms of the expansion:

$$\frac{\partial(\delta\Phi)}{\partial\tau_t} = \tilde{k}\nabla^2(\delta\Phi) + \left(\tilde{p}\,\boldsymbol{exp}\left\{-\frac{\alpha_a}{\Phi_s}\right\}\boldsymbol{exp}\left(-\frac{r^2}{r_0^2}\right)\frac{\alpha_a}{\Phi_s^2} - 1\right)\delta\Phi .$$

(4)

Quantitative analysis of the stability problem of the solutions of equation (4) with respect to perturbations of the form [17]

$$\delta\Phi = A\left(\frac{r}{r_0}\right)^m \boldsymbol{exp}\left(-\frac{r^2}{r_0^2}\right)\boldsymbol{cos}(m\phi)\,\boldsymbol{exp}\left\{\int_0^\tau \lambda dt\right\},$$

where m is an integer, maximum of function $\left(r/r_0\right)^m \boldsymbol{exp}\left(-r^2/r_0^2\right)$ is at a point $r_{max} = r_0\left(m/2\right)^{1/2}$, leads with an approximation $r/r_0 \ll m$ to the dispersion dependence:

$$\lambda(k, \tilde{p}) = \tilde{p}/\tilde{p}_c - 1 - \tilde{k}k^2 ,$$
$$\tilde{p}_c = \frac{\Phi_s^2}{\alpha_a}\boldsymbol{exp}\{\alpha_a/\Phi_s\} ,\ k^2 = \frac{4(m+1)}{r_0^2} . \qquad (5)$$

It is seen that $\lambda(k, \tilde{p})$ depends on the density of radiation in such a way that there is a certain value $\tilde{p} = \tilde{p}_c$ ($\tilde{p}_c$ is the threshold of thermal instability), from which, in the spectrum of perturbations, unstable modes arise. There is an interval of values of the wave vector k that satisfies the inequalities $k^2 < k_c^2 = \varepsilon/\ \tilde{k}$ $\left(\varepsilon = (\tilde{p} - \tilde{p}_c)/\tilde{p}_c\right)$, where $\lambda(k, p) > 0$ and the quasi-stationary solutions Φ_s are unstable with respect to the fluctuations with the wave vector from the specified interval. It is shown that the homogeneous distribution of the temperature field along the beam section at the power density of the radiation $P > P_c$:

$$P_c = \frac{U\tilde{p}_c}{\eta_h T_a} ,\ \tilde{p}_c = \frac{\Phi_s^2}{\alpha_a}\boldsymbol{exp}\{\alpha_a/\Phi_s\} ,\ \Phi_s = \frac{T_s}{T_a}$$

becomes unstable and a structure with a spatially inhomogeneous temperature field profile is formed. The characteristic spatial scale of heterogeneity L_c and the number of rays m, which is determined by the formed self-organized structure, are equal to

$$L_c / r_0 = 2\pi / k_c = 2\pi\Big/\sqrt{\varepsilon/\tilde{k}} ,\ m = (k_c^2 r_0^2 - 1)/4 ,$$

and depend on the radiation power

$$\tilde{p} \quad \left(\varepsilon = (\tilde{p} - \tilde{p}_c)/\tilde{p}_c\right),\ \tilde{k} = \frac{\chi \cdot d_\gamma}{\eta_h r_0^2}).$$

2.2 Mechanism of the Self-organized Structure Formation

This means that the homogeneous distribution of the temperature field along the beam section becomes unstable: at $\tilde{p} > \tilde{p}_c$, ordered structure with a spatially inhomogeneous profile of the temperature field is formed (Fig. 1). Lifetime of the formed self-organized structure is $\tau_{life} = 1/\lambda(\tilde{k}, \tilde{p})$. Short-wave modes, for which $\lambda(k, p) < 0$, quickly fade. For such disturbances, an additional increase in the absorption cross-section due to the temperature at absorption is compensated by the heat dissipation. Thus, changing $\tilde{p}$, it is possible to control the position and shape of the temperature distribution on the plane $z = 0$ between the substrate and the amorphous layer (Fig. 1).

The physical picture of the thermal instability is that heat withdrawal through heat transfer with an amorphous layer and thermal conductivity is not able to compensate the growth of the absorbed energy due to an increase in absorption cross-section with increasing temperature. Threshold values of intensity $\tilde{p}$ and temperature Φ are determined from the condition of equality of the rates of growth of the absorbed energy and its heat removal [14]. Stabilization of thermal instability occurs at the expense of self-regulating mechanisms, which determine the saturation of the absorption non-linearity and equalization of the growth rates of the absorbed energy and heat withdrawal. We note that self-organized modes with a non-uniform temperature distribution combine high intensity of thermal processes with resistance to perturbations, and the structure of the system is stored and maintained due to the joint action of absorption nonlinearity and heat removal over time τ_{life}.

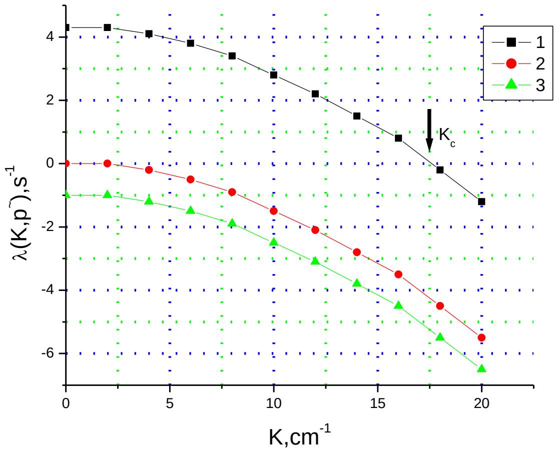

Fig. 1 – Increment of the attenuation $\lambda(k, \tilde{p})$ at different power levels of radiation ($1 - \ \tilde{p} = 0.05 < \tilde{p}_c$, $2 - \ \tilde{p} = \tilde{p}_c = 1.8$, $3 - \ \tilde{p} = 10 > \tilde{p}_c$)

In addition to the above-mentioned formation of a self-organized structure due to thermal instabilities under the influence of IR radiation on the system "substrate-layer", formation of stationary temperature fields determined by bifurcation dependence (3) is possible. In this case, the profile of the temperature field, in addition to the dependence on $\tilde{p}$, is determined by the ratio between the duration of exposure t_p and the lifetime of the self-organized structure τ_{life}.

We analyze the dependence of the threshold density of instability power on the duration of exposure t_p, taking into account the kinetics of the development of thermal instability (Fig. 2). Consider the case where the laser exposure time t_p is much longer than the temperature change due to heat conduction $t_\chi = \rho C r_0^2/(3\chi)$ and heat transfer $t_\eta = \rho C d_\gamma/\eta_h$ that is $t_p \gg t_\chi$, t_η. A characteristic time interval of the evolution of instability t_i is determined by the magnitude $1/\lambda(k,\tilde{p})$ for long-wave disturbances $t_i = (\tilde{p}/\tilde{p}_c - 1)^{-1}$ and correlates with the lifetime of the self-organized structure τ_{life}. As can be seen, the time of development of thermal instability depends on the density of the radiation power, decreases with the growth of the latter, and near the threshold of instability $\tilde{p}_c$ is significant. Therefore, in the region $\tilde{p} \geq \tilde{p}_c$ equation (2) at $\tilde{k} << 1$ and $\tilde{k} << \tilde{p}$ which is realized with $\chi d_\gamma << \eta_h r_0^2$ is reduced to a quasi-stationary one. In this case, the rate of change in the heat flow $Q(T,T_0) = \int_{T_0}^{T} \left(\eta_h / d_\gamma \right) dT$ by the boundary with an amorphous layer is determined by heat transfer with a characteristic time t_η and the amount of energy absorbed by the radiation over time t_p:

$$\frac{dQ(T,T_0)}{dt} = -\frac{Q(T,T_0)}{t_\eta} + \frac{U\gamma}{t_p} p \,. \qquad (6)$$

Thus, from the equation of thermal balance

$$\frac{U\gamma}{t_p} = \frac{Q(T_c,T_0)}{t_\eta p_c}\,,\quad Q(T_c,T_0) = \int_{T_0}^{T_c} \frac{\eta_h}{d_\gamma} dT\,,$$

we get it

$$\frac{dQ(T,T_0)}{dt} = -\frac{Q(T,T_0)}{t_\eta} + \frac{pQ(T_c,T_0)}{p_c t_\eta}\,. \qquad (7)$$

From equation (7), we obtain the dependence of the threshold density of the power of instability from t_p:

$$p = p_c \left(1 - \boldsymbol{exp}\left\{-\alpha_a \cdot t_p / 2t_\eta\right\}\right)^{-1}. \qquad (8)$$

At the density of radiation power $p < p_c$ during the period of laser radiation, a steady-state temperature distribution is established – the temperature of the "substrate-layer" system monotonically increases with growth p and t_p, repeating the spatial beam profile. In the region $p \geq p_c$, the temperature of the substrate sharply increases, and the time of establishing a stationary temperature field, at which saturation nonlinearity of absorption is observed, is determined t_p. At $t_p \leq t_{0p}$ (t_{0p} is the threshold value t_p, which is calculated at the given power of radiation p by the formula (8)), thermal instability is not realized and there is a continuous increase in the temperature of the amorphous layer with increasing exposure. At $t_{0p} < t_p < t_i$, τ_{life}, the establishment of a stationary temperature field at the boundary of the separation of media is impossible, since the rate of growth of energy in absorption significantly exceeds the rate of its heat removal, and in the region $2\pi/\sqrt{\varepsilon/\tilde{k}}$, homogeneous temperature distribution with maximum at $r = 0$ becomes unstable. In a highly supercritical domain $(\tilde{p} >> \tilde{p}_c)$, $t_{0p}/t_\eta \approx \tilde{p}/\tilde{p}_c$ and $t_i/t_\eta \approx \tilde{p}/\tilde{p}_c$, and therefore $(t_{0p} - t_p) \to 0$. In a weakly supercritical region $\left((\tilde{p} - \tilde{p}_c)/\tilde{p}_c << 1\right)$, we have $t_i/t_\eta \approx \tilde{p}_c/(\tilde{p} - \tilde{p}_c)$, $t_{0p}/t_\eta \approx \boldsymbol{ln}\{\tilde{p}_c/(\tilde{p} - \tilde{p}_c)\}$ and $t_{0p} < t_i$. Consequently, the above situation is realized only in a slightly supercritical region (Fig. 2, Fig. 3).

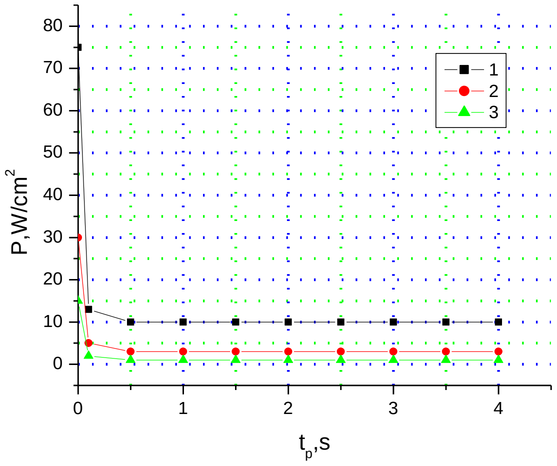

Fig. 2 – Dependence of the threshold density of the power of IR radiation on the duration of radiation t_p ($1 - \alpha_a = 0.6$, $2 - \alpha_a = 0.7$, $3 - \alpha_a = 0.8$)

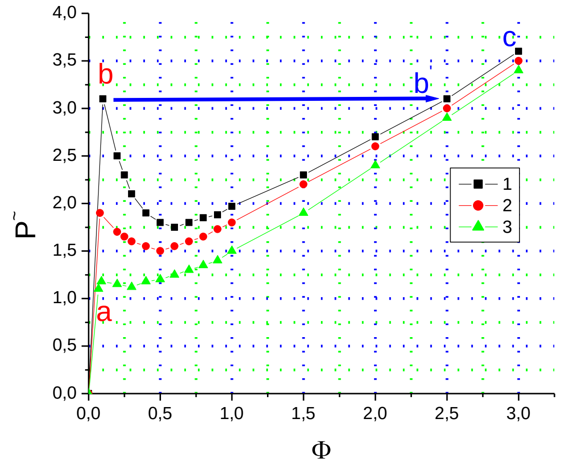

Fig. 3 – Bifurcation diagram and thermal instabilities in the plane $\{\tilde{p}, \Phi\}$ ($1 - \alpha_a = 0.6$, $2 - \alpha_a = 0.7$, $3 - \alpha_a = 0.8$)

When $t_p > t_i$, τ_{life}, there is a saturation of absorption nonlinearity during the time of laser radiation, the stationary temperature mode is set and the temperature profile again corresponds to the spatial distribution of the beam.

Dependence of the threshold power density of the radiation on the duration of the pulse and the bifurcation diagram in the plane $\{\tilde{p},\Phi\}$ are shown in Fig. 3. As follows from the bifurcation diagram $\{\tilde{p},\Phi\}$, with an increase in the power density of the radiation $\tilde{p}$ during the time of laser radiation, a stationary temperature distribution is established (Fig. 3, region ***ab***). The threshold density $\tilde{p}_c$ of laser radiation is determined by the maximum of the dependence $\tilde{p}(\Phi)$ (Fig. 3).

Since

$$\frac{d\tilde{p}}{d\Phi} = e^{\alpha_a/\Phi_s} - (\Phi_s - \Phi_0) e^{\alpha_a/\Phi_s} \left(\frac{\alpha_a}{\Phi_s^2}\right),$$

then to determine the extreme value we obtain the equation

$$\frac{\Phi_s^2}{\alpha_a} - (\Phi_s - \Phi_0) = 0 .$$

Consequently, the threshold value of the power density of the radiation is determined by

$$\tilde{p}_c = (\Phi_s - \Phi_0) e^{\alpha_a/\Phi_s} = \left(\frac{\alpha_a}{\Phi_s^2}\right) e^{\alpha_a/\Phi_s}$$

and coincides with the expression (5). If $\tilde{p} > \tilde{p}_c$, there is a significant increase in temperature (Fig. 3, region ***bb'***). The temperature of the substrate increases sharply, stabilizing with saturation non-linearity of absorption (Fig. 3, region ***b'c)***. The region ***bb'***, which corresponds to the thermal instability of the system, is characterized by the time of its development that is, sometimes attains the value of the stationary temperature field.

3. THERMAL INSTABILITY AND CHANGE OF OPTICAL PARAMETERS FOR NON-CRYSTALLINE THIN FILMS OF GE-AS-TE (S, SE) SYSTEMS

3.1 Results: Bifurcation Diagram

The proposed model makes it possible to investigate the instability and formation of self-organized structures under the action of laser irradiation on layers of amorphous condensates of the Ge-As-Te (S, Se) system. Investigations of the influence of laser radiation have been carried out (wavelengths $\Lambda = 1060$ nm, power density of radiation $P = 3$-10 W/cm^2 and the duration of the exposure $t_p = 0$-30 s on the system "non-crystalline layer + substrate" give an opportunity to experimentally confirm the mechanism of formation of thermal self-organized structures. As an absorption medium of laser radiation, plates of mica or quartz glass were used, but as amophonic layer – layers on the basis of teluro-thick chalcogenide glasses having high crystallization ability. The structural changes in the layer that occur during irradiation are recorded due to a change in the optical density in the visible region of the spectrum $\Lambda = 1060$ nm. The parameters of quartz and mica substrates [1, 10, 16], on which amorphous condensates are applied Ge-As-Te (S, Se), are presented in Table 1.

Layers of amorphous condensates Ge-As-Te are obtained with a thickness of 10-20 nm by the method of discrete thermal sputtering in vacuum $\approx 10^{-4}$ Pa (the rate of condensation was 5-10 Å/s). Outputs of glasses for obtaining layers are selected from the center and near the boundary of the glass formation region. The studies of the structure of layers for Ge-As-Te (S, Se) system are carried out on an electron microscope [4]. Structural features of condensates before and after annealing were established on the basis of diffraction and microdiffraction analyzes of phase composition.

All freshly prepared layers of the Ge-As-Te (S, Se) system are obtained in an amorphous state with a fine-grained structure with a grain size [4]. With an increase in the content of Te in the studied samples, there is an increase in the size of "grains". Annealing of layers at a temperature causes the crystallization of Te of that trigonal modification. An analysis of experimental results on the crystallization of layers in the Ge-As-Te (S, Se) system caused by IR irradiation, suggests the analogy of the processes occurring in them with the same in heat treatment. Quantitative changes in the optical path in layers of chalcogenide glasses are determined in an interference microscope. As can be seen from the interferogram, with irradiation of a laser in the region, there is an inhomogeneous distribution of optical density and a temperature field in the beam section, different from the Gaussian beam profile (Fig. 4). X-ray structural studies of Ge-As-Te glasses showed that, when annealed in X-ray spectra, peaks are characteristic of crystalline Te and GeTe [4]. Fig. 4 shows the time evolution of D of the amorphous layer $(Ge_{20}Te_{80})_{90}(As_2Te_3)_{10}$ as a function of the distance from the laser beam center of radiation determined in densitographs.

For a given density of radiation power at slight exposures, there is a repeats of the spatial profile of a beam with continuous increase of D in the center. With growth of t_p, the optical density D in the center of the laser beam begins to decrease and then grows again, going to saturation (Fig. 5).

Table 1 – Thermodynamic and optical parameters of quartz and mica substrates

Type	ρ, g·cm^{-3}	C, J·g^{-1}·K^{-1}	d_γ, nm	χ, W·cm^{-1}K^{-1}	η_h, W·cm^{-2}K^{-1}	T_a, K	d, cm
quartz glass	2.4	0.71	99	$0.84 \cdot 10^{-2}$	$1.2 \cdot 10^{-2}$	2100	0.2
mica	2.5	0.83	94	$0.9 \cdot 10^{-2}$	$2 \cdot 10^{-2}$	1800	0.08

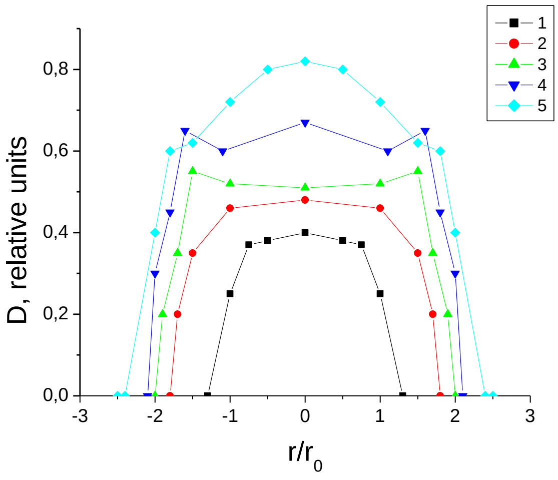

Fig. 4 – Relative change in optical density of $(Ge_{15}Te_{85})_{90}(AsTe)_{10}$ layers under the action of CO_2-laser ($I = 0.3$ W, 1 – $t_p = 0.1$ s, 2 – $t_p = 0.2$ s, 3 – $t_p = 0.3$ s, 4 – $t_p = 0.5$ s, 5 – $t_p = 0.7$ s)

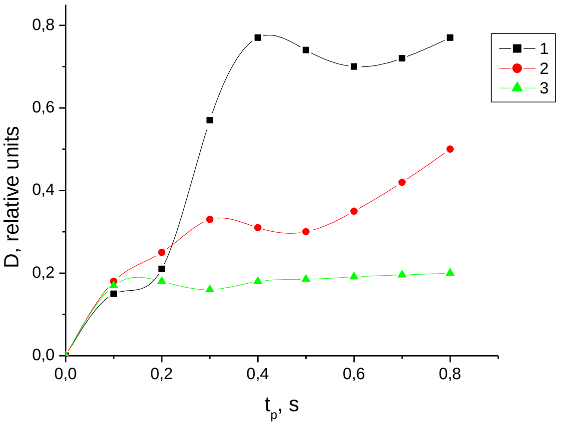

Fig. 5 – Relative change in optical density of Ge-As-Te layers as a function of the duration of laser irradiation: 1 – $(Ge_{15}Te_{85})_{90}(AsTe)_{10}$, 2 – $(Ge_{20}Te_{80})_{90}(As_2Te_3)_{10}$ and 3 – $(Ge_{15}Te_{85})_{88}(AsTe)_{12}$

The recording of the action of radiation in the conditions of thermal heating of the "amorphous layer + substrate" system is characterized by an increase in the sensitivity of the material and a decrease in the threshold density, in which the effect of a singular change in optical density is observed. At irradiation or annealing of amorphous condensates, there is a separation of the crystalline phase, which causes the change in their optical density. The conducted electron microscopic studies of irradiated layers indicate their crystallization under the action of radiation. Thus, the above distribution D reflects the spatial temperature profile in the recording process. The effect of a singular change of D can be considered as the result of the development of thermal instability and the formation of self-organized structures [6, 15]. The estimates of parameters of thermoinduced instabilities of amorphous condensates of the Ge-As-Te system according to the formulas (3)-(8) are carried out. Calculations for

$$\tilde{k} = \frac{\chi d_\gamma}{\eta_h r_o^2} \approx 5\cdot 10^{-3} << 1$$

and values of time intervals of heat dissipation $t_\eta \approx 5\cdot10^{-3}$-$3\cdot10^{-2}$ s and thermal conductivity $t_\chi \approx 10^{-2}$-$5\cdot10^{-2}$ s substantiate the use of a quasi-stationary approximation (6). The dependence of the threshold density of radiation power on the duration of exposure and the bifurcation diagram in the plane $\{P, T\}$ for an amorphous $(Ge_{20}Te_{80})_{90}(As_2Te_3)_{10}$ is shown in Fig. 6. As follows from the bifurcation diagram $\{P, T\}$, with the increase of the power density of the radiation P during the time of laser radiation, a stationary temperature distribution is established (Fig. 6, the region ***ab***). At $P > P_c = 7.6$-8.5 W/cm², where P_c is a power density threshold, which is determined by maximum dependence $P(T)$ (Fig. 6), there is a significant increase in temperature (Fig. 6, region ***bb'***). The region ***bb'***, which corresponds to the thermal instability of the system, is characterized by the time of its development and the achievement of the value of the stationary temperature field $t_i \approx 10^{-2}$-$4\cdot10^{-1}$ s. Characteristic parameters of the beam cross section $r_0 = 0.11$ cm and amorphous condensates of the Ge-As-Te system: $U = 0.60$, $L_a = 986$ J/g, $E_a = 0.52$ kcal/mol. For $P = 7.8$ W/cm² ($I = 0.3$ W, $r_0 = 0.11$ cm) and $P_c = 7.6$ W/cm² threshold duration of the pulse, at which instability develops, is of the order of 0.35÷0.4 s, number of rays $m = 0$, which correlates with exposures shown in Fig. 6 in the range of nonmonotonic behavior of D. Characteristic values of the spatial scale of heterogeneity of the temperature profile $L_c \approx (0.3$-$0.5)\cdot r_0$, the lifetime of the self-organized structure $t_{life} \approx 10^{-1}$÷10^{-2} s and are consistent with experimental data. Radial-ring structure with the number of rays $m \geq 3$ can be formed with irradiation power densities $P \geq 18.6$ W/cm².

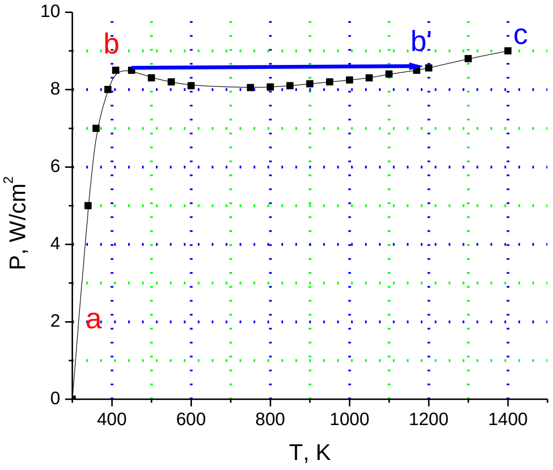

Fig. 6 – Bifurcation diagram in plane $\{P, T\}$ for amorphous film $(Ge_{15}Te_{85})_{88}(AsTe)_{12}$

Consequently, the considered laws of the effect of IR radiation on amorphous layers can be analyzed in the framework of the approach that takes into account the feedback, which is carried out through the nonlinear dependence of absorption on the intensity of radiation, and allows us to reveal the physical picture of the dynamics of a strongly non-equilibrium system. This effect is observed not only in the layers of amorphous condensates Ge-As-Te, but also in multilayered structures based on As-S (Se) at significantly lower radiation intensities. It should be noted that the development of recording environments on the basis of amorphous condensates of Ge-As-Te, Ge-Te, As-S (Se) systems causes interest and is applied in connection with the problems of laser radiation diagnostics, the ability to create archival memory and video discs on their basis [8-11]. Similar effects were observed by the authors in investigating the effect of a CO_2-laser on layers based on glassy As_2S_3 on oxide glass substrates. Distinctive features in this case are related to the fact that the melting-crystallization temperature of the As-S (Se) layers ($T_m \approx$ 500-550 K) is lower than that of the amorphous condensates on the basis of Ge-As-Te, and, consequently, lower threshold radiation for power densities should be expected P_c. As numerical calculations show, the threshold power for amorphous layers based on As_2S_3, the radiation power density is $P_c \approx$ (1.5-3) W/cm^2.

3.2 Discussion

The above principles of the level formation of the ordered self-organized structures in thin films can be applied to the development of their fractality. Another extremely relevant and interesting aspect of the research in this paper is that the hierarchy of time scales for the development of thermo-, laser induced instabilities has a fractal nature [6, 19] and suggests the possibility of forming the hyper sensibility of self-organized structures (Table 2). Fractality is manifested through the lifetime of self-organized structures, which acts as one of the iterative parameters (this issue will be discussed in more detail in the future).

Table 2 – The hierarchy of time scale development of instability and the formation of a fractal structure

Time of laser irradiation	t_p, $t_p = \left(0 \div 30 \quad s\right)$
Time of temperature change due to thermal conductivity	$t_\chi = \rho C r_0^2/(3\chi)$, $t_\chi \approx (10^{-2} \div 5 \cdot 10^{-2})s$
Time of temperature change due to heat transfer	$t_\eta = \rho C d_\gamma/\eta_h$, $t_\eta \approx (5 \cdot 10^{-3} \div 3 \cdot 10^{-2})s$
A characteristic time interval of the evolution of instability	$t_i = \left(\tilde{p}/\tilde{p}_c - 1\right)^{-1}$, $t_i \approx 10^{-2} \div 4 \cdot 10^{-1} \quad s$
Lifetime of the self-organized structures	τ_{life}, $\tau_{life} \approx 10^{-1} \div 10^{-2}s$
$\tau_{life} \propto \quad t_i \propto \quad t_p >> t_\chi$, t_η	

4. CONCLUSIONS

A mechanism of thermal instability and the formation of self-organized structures under the action of continuous IR radiation of a CO_2-laser on amorphous condensates of Ge-As-Te (S, Se) systems applied to quartz or mica substrates is proposed, and its analysis is based on experimental studies. It is shown that the uniform distribution of the temperature field along the beam section at the power density of the radiation $P > P_c$ (P_c = 1.8÷8.5 W/cm^2) becomes unstable and a radial-ring structure with a spatially inhomogeneous profile of the temperature field with a period $L_c \approx (0.3 \div 0.5) \cdot r_0$ (r_0 is an effective radius of the beam) is formed. Dependence of the threshold density of the power P_c of IR radiation of a CO_2-laser on the duration of exposure is established. A bifurcation diagram in a plane *{radiation power (P), temperature (T)}* that describes the singular behavior of optical density depending on the duration of exposure to radiation is constructed. It is shown that the stabilization of thermal instability and the formation of a nonuniform temperature profile are carried out at the expense of self-regulating mechanisms, which lead to saturation of the absorption nonlinearity and equalization of the growth rate of the absorbed energy and heat transfer into the amorphous layer. One of the ways of forming fractality of self-organized structures is analyzed.

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REFERENCES

1. J.L. Adam, X. Zhang, *Chalcogenide Glasses: Preparation, Properties and Applications* (London: Elsevier Science: 2014).
2. A.V. Dalekorej, V.P. Ivanitsky, V.S. Kovtunenko, M.V. Stoika, *J. Nano- Electron. Phys.* **9** No5, 05020(6pp) (2017).
3. R. Svoboda, J. Malek, *J. Non-Cryst. Sol.* **419** No1, 39 (2015).
4. I.I. Turjanitca, M.I. Mar'yan, *Ukr. Phys. J.* **37** No2, 256 (1992).
5. A.N. Kuryliuk, K.V. Maliutiak, V.V. Kuryliuk, *J. Nano-Electron. Phys.* **11** No 2, 02001(5pp) (2019).
6. M. Mar'yan, N. Yurkovych, *Self-organized structures in non-crystalline solids and other systems: Methods, concepts and applications to the information technology* (Saarbruecken: LAP Lambert Academic Publishing: 2019).
7. M.I. Mar'yan, A.A. Kikineshy, A.A. Misak, *Phil. Mag. B: Phys. Condens. Mat.* **68** No 5, 689 (1993).
8. M. Popescu, *J. Opt. Adv. Mat.* **5** No 5, 1059 (2003).
9. K. Tanaka, *J. Non-Cryst. Sol.* **336** No 5, 21 (2004).
10. J. Teteris, M. Reinfelde, *J. Opt. Adv. Mat.* **5** No 5, 1355 (2003).
11. M.F. Thorpe, D.J. Jacobs, M.V. Chubynsky, J.C. Phillips, *J. Non-Cryst. Sol.* **266** No 5, 859 (2000).
12. M. Mar'yan, A. Szasz, P. Szendro, A. Kikineshy, *J. Non-Cryst. Sol.* **351** No 2, 189 (2005).
13. N. Yurkovych, M. Mar'yan, V. Seben, *Sem. Phys. Quant. Electr. Opt.* **21** No 4, 365 (2018).
14. M.I. Mar'yan, *Sc. Her. Uzh. Univ. Ser. Phys.* **3**, 114 (1998).

15. M. Mar'yan, N. Yurkovych, *XVII International Freik Conference on the Physics and Technology of Thin Films and Nanosystems (ICPTTFN- XVII),* 252 (Ivano-Frankivsk: Publisher Vasyl Stefanyk Precarpathian National University: 2019); N. Yurkovych, M. Mar'yan, V. Seben, ISSN 2519-8297, 361 (Ivano-Frankivsk: Publisher Vasyl Stefanyk Precarpathian National University: 2019).
16. N.F. Mott, E.A. Davis, *Electronic Processes in Non-Crystalline Materials* (Oxford: Oxford University Press: 2012).
17. P. Shuster, *Stochasticity in Processes: Fundamentals and Applications to Chemistry and Biology* (Berlin: Springer: 2016).
18. E. Frehland, *Synergetics, From Microscopic to Macroscopic Order* (New York: Springer: 2011).
19. M. Mar'yan, V. Seben, N. Yurkovych, *Synergetics and Fractality in Science Education* (Presov: University of Presov Publishing: 2018).

Формування та Моделювання Нанорозмірних Рівнів Самоорганізованих Структур в Некристалічних Тонких Плівках Систем Ge-As-Te(S,Se)

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Запропонована модель теплових нестійкостей і формування самоорганізованих структур при дії неперервного інфрачервоного випромінювання CO_2-лазера на аморфні конденсати систем Ge-As-Te (S, Se), нанесені на підкладки з кварцу або слюди, та проведено її аналіз на основі експериментальних досліджень. Показано, що однорідний розподіл температурного поля вздовж перерізу пучка при густинах потужності випромінювання $P > P_c$ ($P_c = 1.8$-8.5 Вт/см^2) стає нестійким і формується радіально-кільцева структура з просторово-неоднорідним профілем температурного поля. Встановлено, що біфуркація теплової нестабільності та утворення нерівномірного температурного профілю здійснюється за рахунок саморегулюючих механізмів, що призводять до насичення нелінійності поглинання та вирівнювання швидкості росту поглиненої енергії шляхом передачі тепла в аморфний шар. Досліджена залежність порогової густини потужності інфрачервоного випромінювання формування впорядкованих радіально-кільцевих структур від тривалості опромінення. Побудована біфуркаційна діаграма в площині *{потужність випромінювання (P), температура (T)}*, яка описує сингулярну поведінку оптичної густини в залежності від тривалості експозиції опромінення. Показано, що радіально-кільцева структура з числом променів $m \geq 3$ формується для аморфних конденсатів систем Ge-As-Te (S, Se) при густинах потужності опромінення $P \geq 18.6$ Вт/см^2, що узгоджується з експериментальними даними. Проаналізовані особливості поведінки самоорганізованих структур в тонкоплівкових шарах, а саме чутливість системи до технологічних умов одержання, параметрів електромагнітного випромінювання та можливість реалізації ефекту метелика. Розглянуто способи формування та реалізації фрактальності за нанорозмірними рівнями структурування та тривалості життя самоорганізованих структур у некристалічних матеріалах систем Ge-As-Te (S, Se).

Ключові слова: Ефект інфрачервоного опромінення, Фрактальність, Некристалічні тонкі плівки, Самоорганізовані структури, Синергетика.