BISTABILITY THERMOINDUCTION AND DISSIPATIVE STRUCTURE FORMATION IN THE NON-CRYSTALLINE MATERIALS

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The effect of the powerful laser light (λ = 10.6 μm) on the amorphous films has been studied. The model which takes into account non-linear temperature dependence of the absorptivity and nonstationarity of the heat removal process is considered. It describes irreversible changes under laser irradiation and effect of nonmonotonous changes in optical density of the amorphous film depending on the pulse length.

1. Introduction

Laser radiation interaction with different-type non-crystalline solids produces the photo-induced change of their optical [1-3], physico-chemical [4] and other properties. These processes have been fairly well studied in the chalcogenide vitreous semiconductors [1-4], where they are most effective under the exposure of the light quants of $\hbar\omega \leq E_g$ energy. Interest in the studies of the photo-induced transformations in such materials, besides the practical application, is due to the fact that the non-crystalline media produced in the strongly non-equilibrium conditions are open over the energy and mass exchange. Due to this the self-organizing phenomena in the non-crystalline structures under illumination appear to be diverse, and qualitatively new stationary and non-stationary structures have the possibility to occur. A series of experimental data on the studies of the dynamics of the optical parameter variations and relaxation phenomena at the photo-thermal excitation [5-6] indicate the specific features of wide-spectrum $(\lambda = 0.3 - 15 \mu m)$ laser radiation interactions. These data can be analysed, as shown below, from the viewpoint of the photo-induced instabilities and the dissipative structure formation. The thermal and electron mechanisms of the laser radiation action is defined primarily by the energy density.

At moderate energy densities ($\leq 1 W/cm^2$) predominant is the contribution related to the change of the state of electron subsystem and subsequent vibrational spectrum transformation. At higher energy densities $\left(\geq 10 W/cm^2\right)$ the photocrystallisation processes due to the thermal component of the laser radiation action are observed for many non-crystalline materials [6].

Production of the spatio-temporal structures and thermal instabilities with the bistable behaviour under the influence of the ultra-short nanosecond and picosecond pulses or the powerful continuous radiation on the semiconductor layers and biostructures is one of the possible examples of the systems which allow the formation of the dissipative structures far away from the equilibrium state [7-9]. The mechanism of the production of these structures is based on the linear interaction of radiation with the substance when the reverse influence of the medium on the radiation absorption occurs, and the stationary non- equilibrium mode is possible possessing the spatial and time characteristics which differ qualitatively from those for the linear energy transfer mechanism. In this section, we shall study the development and stabilisation of thermal instabilities with simultaneous formation of dissipative structures in the non- crystalline layers under the influence of the continuous IR- radiation.

2. Model of system and thermal instability formation

Consider the model system comprising the cover layer, the non-crystalline material and the substrate. Let the radiation of $q(x)=I(x)/\pi x_0^2$ power with the Gaussian intensity distribution $I(x)=I_0\,exp\{-x^2/x_0^2\}$ over the beam cross section (x_0 is the effective beam radius) and with the pulse duration of t, whose intensity decreases along the z-axis, be absorbed completely by the system volume. The peculiarities of the behaviour of the IR- transparent layers of non-crystalline semiconductors [10] are defined by the substrates, which absorb the radiation of a given wavelength, and the heat removal mechanism. In the IR-region of the spectrum, the strong temperature dependence of the absorption cross section appears to be essential. The absorbing capacity can be approximated by the expression $\beta=\beta_0 exp\{-\alpha T_a/T\}$, where α is the non-linearity parameter, β_0, T_a - constants. A considerable increase of the absorption and the rise of the reverse influence of the medium on the radiation take place with the radiation energy density increase. The self-consistent increase of the absorption at the presence of the feedback between the radiation and the substance as well as the occurrence of the non- stationary temperature mode at the non-crystalline boundary may result in the appearance and development of thermal instability self-organisation.

In order to study the formation of the structures due to the thermal instabilities, consider the dynamics of the temperature variation in the "substrate + layer" system. This problem is described by the non-linear differential equation which accounts for heat generation, propagation and transfer:

$$\rho C\frac{\partial T}{\partial t}=div(\kappa grad T)+G(T)-Q(T). \quad (1)$$

Here C, ρ and κ are the heat capacity, density and heat conduction, respectively; $G(T)=\beta U q\delta(z)f(t)$ is the heat source; U is the optical transmission of the layer at the λ wavelength which allows for the power variation at the absorption; $Q(T)=\int_{T_0}^{T}\frac{\eta(\xi)d\xi}{d}$ is the heat exchange with the non-crystalline layer (η being the heat exchange constant, T_0 being the initial temperature),

$$\delta(z)=\begin{cases}1, & z\le d,\\ 0, & z>d,\end{cases}\qquad f(t)=\begin{cases}1, & t\le t,\\ 0, & t>t.\end{cases}$$

The boundary conditions are:

$$T(x,z,t)\big|_{z=0}=T_0,\ \left.\frac{\partial T(x,z,t)}{\partial z}\right|_{z=0}=0 \quad .(2)$$

To solve the equation (1) and analyse the obtained results it is convenient to use the dimensionless values: $\Phi=\frac{T}{T_a}$, $\Phi_0=\frac{T_0}{T_a}$, $\tilde{q}=\frac{Uq\delta(z)}{\eta T_a}$, $\tau=\frac{\eta t}{\rho C d}$, $\mu=\frac{\kappa d}{\eta x_0^2}$ in terms of which the equation (1) attains the form:

$$\frac{\partial\Phi}{\partial\tau}=\mu\nabla^2\Phi+\tilde{q}\,exp\left\{-\frac{\alpha}{\Phi}\right\}f(\tau)-\Phi+\Phi_0 \quad (3)$$

Here $\nabla^2\Phi$ is the Laplace operator in the cylindrical coordinate frame. The stationary temperature field which specifies the bifurcation diagram in the $\{\Phi,q\}$ plane is given by the equation:

$$\nabla^2\Phi_s=-\tilde{q}\,exp\left\{-\frac{\alpha}{\Phi}\right\}+\Phi_s-\Phi_0. \quad (4)$$

The configuration of the stationary temperature field depends on the geometry of the two-layer "non-crystalline layer + substrate" system, the heat source density and the heat removal value. The system possesses the bistability over the $\tilde{q}$ parameter related to the radiation intensity. Depending on $\tilde{q}$, one or three stationary states do exist differing in the reduced temperature Φ. The type of the singular points in the bifurcation diagram $\{\Phi,\tilde{q}\}$ and their stability are studied by expanding Φ over the small deviations from the stationary solutions $\Phi=\Phi_s+\delta\Phi$, where Φ_s is a root of equation (4), $\delta\Phi$ is a variation of Φ [5]. Substituting $\Phi=\Phi_s+\delta\Phi$ into equation (3)

and holding only the terms linear over $\delta\Phi$, we obtain:

$$\frac{\partial(\delta\Phi)}{\partial\tau}=\mu\nabla^2(\delta\Phi)+\left(\frac{\tilde{q}\Phi_s^2}{\alpha}exp\left\{\frac{\alpha}{\Phi_s}\right\}+1\right)\delta\Phi. \qquad (5)$$

The quantitative analysis of the stability of the solutions of equation (5) with respect to perturbations of the form:

$$\delta\Phi=J_m(P\xi)cos(m\varphi)exp\left\{\int_0^\tau\gamma dt\right\}, \qquad (6)$$

where $J_m(P\xi)$ is the first-order Bessel function over m, leads us to the following dispersion dependence:

$$\gamma(P)=\tilde{q}/\tilde{q}_c-1-\mu P^2,\ \tilde{q}_c=\frac{\Phi_s^2}{\alpha}exp\{\alpha/\Phi_s\}. \qquad (7)$$

It is seen that $\gamma(P)$ depends on the radiation density in such a way that a certain value of $\tilde{q}=\tilde{q}_c$ does exist ($\tilde{q}_c$ being the thermal instability threshold), starting with which the unstable modes appear in the perturbation spectrum. One can distinguish the interval of the wave vector values P which obeys the inequality $P^2<P_c^2=\varepsilon/\mu$ $\left(\varepsilon=(\tilde{q}-\tilde{q}_c)/\tilde{q}_c\right)$, where $\gamma(P)>0$ and the quasi-stationary solution Φ_s is unstable with respect to the fluctuation with the wave vector taken from the given interval. A specific space scale of the instability zone which defines the formation of the dissipative structure is $2\pi/P_c=2\pi/\sqrt{\varepsilon/\mu}$ and depends on $\tilde{q}$. The short-wave modes for which $\gamma(P)<0$ and the modulus is rather large, are dampened rapidly. For such perturbations the additional increase of the absorption cross section caused by the temperature is compensated by the heat removal.

The physical pattern of the thermal instability resides in that the heat removal through the non-crystalline layer boundary and the heat transfer fail to compensate the rise of the energy absorbed due to the temperature increase of the absorption cross section. The threshold intensity $\tilde{q}$ and the temperature Φ are defined provided that the rate of the absorbed energy increase is equal to that of heat removal. Thermal instability stabilisation occurs at the cost of the self-regulated mechanisms resulting in the saturation of the absorption non-linearity which governs the equalisation of the rate of the absorbed energy increase and that of heat removal. Note that the self-organising modes combine the high intensity of thermal processes with the perturbation resistance, while the form of the structure is retained due to the joint action of the non-linear absorption and heat removal.

Let us analyse the threshold power density of the instability as a function of t_p taking into account the kinetics of the development of the heat instability. Consider the situation when t_p is much more than the time of the temperature change due to the heat conduction $t_\kappa=\rho Cx_0^2/3\kappa$ and the heat transfer $t_\eta=\rho Cd/\eta$, i.e. $t_p>>t_\kappa$, $t\eta$. A specific time interval of the instability evolution t_i is defined by the $\gamma(P)$ value and for the long-wave perturbation $t_i=(q/q_c-1)^{-1}$. As is seen, the time of the heat instability evolution depends on the radiate power and is decreased with the latter, while close to the threshold q_c is large. Therefore, in the hypercritical region, the evolution equation (1) at $\mu<<1$ is reduced to the quasi-stationary one:

$$\frac{dQ(T,T_0)}{dt}=-\frac{Q(T,T_0)}{t_\eta}+\frac{qQ(T_c,T_0)}{q_c t_\eta}f(t),$$

$$Q(T_c,T_0)=\int_{T_0}^{T_c}\eta(\xi)d\xi. \qquad (8)$$

From equation (8) we obtain the threshold capacity of instability as a function of t:

$$q=q_c\left(1-exp\left\{-\alpha t/2t_\eta\right\}\right)^{-1}. \qquad (9)$$

At the radiate power density $q<q_c$, a stationary temperature distribution is set, and the temperature of the "substrate + layer" system is increased monotonically with q and t_p reproducing the spatial beam profile.

In the hypercritical region, $q>q_c$, the substrate temperature increases sharply, and the time of the stationary temperature field relaxation at which the absorption non-linearity is saturated is defined by t_p. At $t_p>t_{0p}$ (t_{0p} is the threshold value of t_p defined at the given radiate power q by formula (9)), the heat instability does not occur and a continuous increase of the amorphous layer temperature with exposure time is observed. At $t_{0p}<t_p<t_i$, the establishment of the stationary temperature field at the interface is impossible since the energy increase rate at the absorption exceeds considerably the heat removal rate and in the region of the $2\pi/\sqrt{\varepsilon/\mu}$ order the homogeneous temperature distribution with the maximum at $\xi=0$ becomes unstable. In the highly hypercritical region $(q>>q_c)$, $t_{0p}/t_\eta \approx q_c/q$ and $t_i/t_\eta \approx q_c/q$, and, therefore, $(t_{0p}-t_p)\to 0$. In the weakly hypercritical region $((q-q_c)/q_c<<1)$, $t_i/t_\eta \approx q_c/(q-q_c)$ and $t_{0p}/t_\eta \approx ln\{q_c/(q-q_c)\}$ and $t_{0p}<t_i$. Thus, the above situation occurs only in the weakly hypercritical region.

At $t_p>t_i$, a non-linear absorption saturation is observed in a time of laser action resulting in the establishment of a stationary temperature mode, and the temperature profile again corresponds to the spatial distribution of the beam.

3. Thermal instabilities evolution and variation of the optical parameters for certain non-crystalline media

The model considered above allows one to explain the regularities of the IR laser radiation with condensed media. The studies of the influence of the laser radiation (with $\lambda=10.6\mu m$ wavelength, $q\,(3-5W/cm)$ power and $t_p(0-30s)$ pulse duration) on the two-layer "non-crystalline layer + substrate" system [10,11] allow the above mechanism to be confirmed. The mica and quartz plates were used as the laser radiation absorbing medium, while the tellurium-containing chalcogenide vitreous glass-based layers with good crystallisation ability served as the transparent amorphous layer [12]. The structural changes in the layer occurring under radiation were detected by measuring the optical density D in the visible spectral region $(\lambda=0.63\mu m)$ by means of a densitometer.

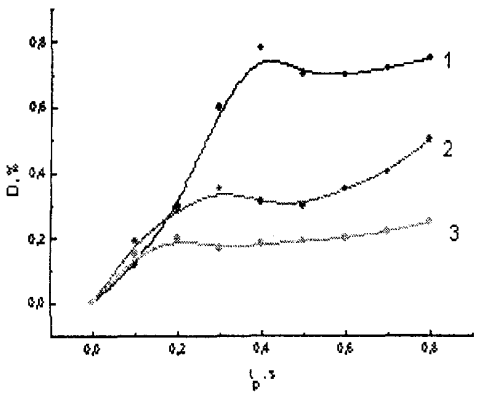

Fig.1. Relative variation of the optical density of the Ge-As-Te layers as a function of the duration of the radiate influence.
1-$(Ge_{15}Te_{85})_{90}(AsTe)_{10}$, 2 - $(Ge_{20}Te_{80})_{90}(As_2Te_3)_{10}$, 3 - $(Ge_{15}Te_{85})_{88}(AsTe)_{12}$.

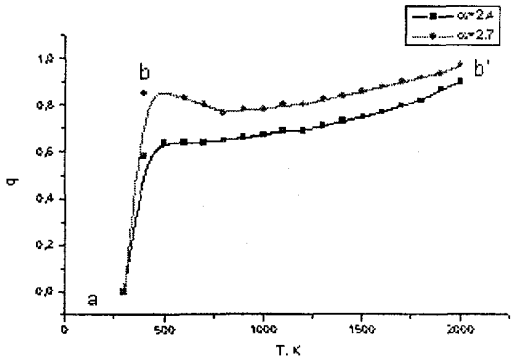

Fig.2. Thermal instability intensity as a function of pulse duration.

The time evolution of D for the $Ge-As-Te$ determined by the densitogram is shown in Fig.1 as the function of the distance from the centre of the beam. For given radiate power at low exposures the spatial profile of the beam is repeated with the continuous increase of D in its centre (see Fig.1). With increasing t, D at first decreases and then again increases tending to saturation. It should be noted that for the amorphous $(Ge_{15}Te_{85})_{88}(AsTe)_{12}$, $(Ge_{15}Te_{85})_{90}(AsTe)_{10}$ layers the thermal instability occurs at the energy densities of about $(1.7-5.2)J/cm^2$, while for the $(Ge_{20}Te_{80})_{90}(As_2Te_3)_{10}$ layer this occurs at $42\,J/cm^2$. The change in the optical density at the centre of the laser spot in the region of the leap for the materials under consideration is about 10% (see Fig. 1). The radiation record in the conditions of thermal heating of the system "substrate + layer" is characterised by the rise of the material sensitivity and the decrease of the threshold density at which the effect of non-monotonic variation of the optical density is observed. The electron-microscopic studies of the irradiated $Ge-As-Te$ layers indicate their radiation-induced crystallisation and, consequently, the above distribution of D reflects the spatial temperature profile during the process of recording. The effect of the non-monotonic variation of D can be treated as the result of the thermal instability evolution.

Let us perform some estimations in accordance with formulae (7)-(9). For the quartz glass substrate t_η, $t_\kappa \cong 10^{-2}s$, $\mu = 0.13$ (here we used the values of $\rho = 25\,g/cm^3$, $C = 0.83\,J/gK$,

$\kappa = 10^{-2}\,W/cmK$,

$\eta = 2\cdot 10^{-2} W/(cm^2K)$,

$d = 0.34\mu m$, $u = 0.69$), $q = 7.66\,W/cm^2$.

The dependence of the threshold radiative power on the pulse duration and the bifurcation diagram in the $\{q,T\}$ plane are Fig.2. As follows from the bifurcation diagram $\{q,T\}$, the temperature of the system at first increases monotonically with increasing radiative power density q (a stationary temperature distribution defined by the diagram at the ab area is established during the laser radiation). At $q > q_c$ (here q_c is a density threshold defined by the maximum in the dependence $q(T)$) a considerable temperature rise occurs (the bb' area). The substrate temperature increases sharply being stabilised at the saturation of the absorption non- linearity (the bc area). The bb' area which corresponds to the thermal instability of the system is characterised by the time of its evolution, i.e. the time which is required to achieve the stationary temperature field (10^{-2} to 10 s for the system under study). For $q = 7.9\,W/cm^2$ ($I = 0.3W$, $x = 0.11cm$), the threshold pulse duration, at which the instability is produced, is about $0.35-0.4s$ and correlates with the exposure times in the interval of the non-monotonic behaviour of D (see Fig.1,2).

Thus, the observed regularities of the IR-radiation influence on the non-crystalline tellurium-containing glass layers are explained within the framework of the feedback approach for the "radiation + substance" system realised by the non-linear temperature behaviour of the absorption and allow one to unravel the physical pattern of evolution of highly non- equilibrium system.

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ТЕРМОІНДУКОВАНА БІСТАБІЛЬНІСТЬ ТА ФОРМУВАННЯ ДИСИПАТИВНИХ СТРУКТУР В НЕКРИСТАЛІЧНИХ МАТЕРІАЛАХ

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Досліджується ефект впливу лазерного випромінювання (λ = 10.6 μm) на аморфні шари Ge-As-Te. Розглянута модель, яка враховує нелінійну температурну залежність коефіцієнта поглинання та нестаціонарні теплові процеси. Описано немонотонну зміну оптичної густини аморфних шарів в залежності від тривалості опромінення.