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To Question of Modeling of Mechanisms of Laser-Induced Destruction in Dielectrics and Semiconductors

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Results of analysis of problem of mechanisms of laser-induced destruction in dielectrics, including polariton models, are discussed. These phenomena may be represented as effects of Relaxed Optics. The influence of dynamic and kinetic factors on these processes is shown. Some analogy of these mechanisms with microscopic mechanism of Cherenkov radiation is analyzed too.

Key words: laser-induced destruction, dielectrics, Relaxed Optics, laser implantation, polariton models, Cherenkov radiation, radiate relaxation, nonradiative relaxation.

Трохимчук П. П. До питання моделювання механізмів лазерно-індукованого руйнування діелектриків та напівпровідників. Наведено результати аналізу проблеми моделювання механізмів лазерно-індукованого руйнування діелектриків, включаючи поляритонні моделі. Ці явища розглянуто як ефекти релаксаційної оптики. Показано, що поряд з динамічними чинниками в цих процесах вирішальну роль можуть відігравати і кінетичні чинники. Проаналізовано певну аналогію цих механізмів з мікроскопічним механізмом Черенковського випромінювання.

Ключові слова: лазерно-індуковане руйнування, діелектрики, релаксаційна оптика, лазерне легування, поляритонні моделі, Черенковське випромінювання, випромінювальна релаксація, безвипромінювальна релаксація.

Трохимчук П. П. К вопросу моделирования механизмов лазерно-индуцируемого разрушения диэлектриков и полупроводников. Приведены результаты анализа проблемы моделирования механизмов лазерно-индуцируемого разрушения диэлектриков, включая поляритонные модели. Эти явления рассмотрены как эффекты релаксационной оптики. Показано, что наряду с динамическими факторами в этих процессах решающую роль могут играть и кинетические факторы. Проанализирована определенная аналогия этих механизмов с микроскопическим механизмом Черенковского излучения.

Ключевые слова: лазерно-индуцируемое разрушение, диэлектрики, релаксационная оптика, лазерное легирование, поляритонные модели, Черенковское излучение, излучательная релаксация, безизлучательная релаксация.

Introduction. A problem of modeling of laser-induced destructions in dielectrics is one of significant problems of modern power laser physics [3, 11–13]. This problem may be resolved with help of modeling laser induced polariton-plasmon effects [4, 9, 10, 13] and it is very important for the research of the creation the damages in dielectrics after laser irradiation with $h\nu \leq, \ll E_g$, where $h\nu$ – photon energy of laser, E_g – a band gap of irradiated dielectrics. This mechanism is basic for the laser destruction of materials with irradiating with picosecond and femtosecond laser pulses [3, 9, 10, 13].

Basic difference between polariton optical phenomena in dielectrics [3, 13] and polariton mechanisms of creation damages in dielectrics [9, 10, 13] is consisted in various mechanisms of relaxations of first-order excitations: radiative relaxation for «optical» case and non-radiating relaxation for «destruction» case.

Analogous effects and models are used for the explanation of microscopic nature of Cherenkov radiation [1, 2, 5, 7, 8].

Therefore the one basic object of this manuscript is investigation of possibility the use the results of modeling of microscopic nature of Cherenkov radiation for the creation microscopic models of the creation laser-induced damages in dielectrics.

Basic results and its discussions. The problems of creation the mechanisms of irreversible interactions light and solid are one of basic problems of Relaxed Optics. Universal polariton model of laser destruction of materials is based on the multiphoton absorption of laser radiation [9, 10].

This problem is connected with idea of saturation of excitation of proper center of light scattering in space and time. It follows from theoretical calculations and experiments that due to multiphoton transitions and avalanche ionization semiconductor undergoes the nonthermal phase transition into the metallic state

with concentrations of nonequilibrium electrons up to $n_e = 10^{21} \text{ cm}^{-3}$ and higher. Plane layers of laser excited dielectric, which have metallic properties, are generated for high level of laser intensity for time $\sim 10^{-14} \text{ s}$.

If for the boundary plasma layer- semiconductor (refractive index n) the condition:

$$\omega_0 < \omega_0 < \omega_p, \quad 1 + n^2 \omega_0^{-2} = \omega_{sp}^2, \quad \omega_p (1 + n^2)^{-1/2} = \omega_{sp}, \quad (1)$$

has place; where ω_0 – a frequency of laser irradiation, ω_{sp} – a frequency of surface plasmon, ω_p – plasmic frequency.

Physical sense of formula (1) is next. Laser frequency must be less as limiting frequency of surface plasmon polariton (SPP) of border dielectric – plasmic layer (surface active matter). Realization of this case without dissipation ($\gamma_{e-e} = 0$) is represented in Fig.1. γ_{e-e} is frequency of electron-electron collisions.

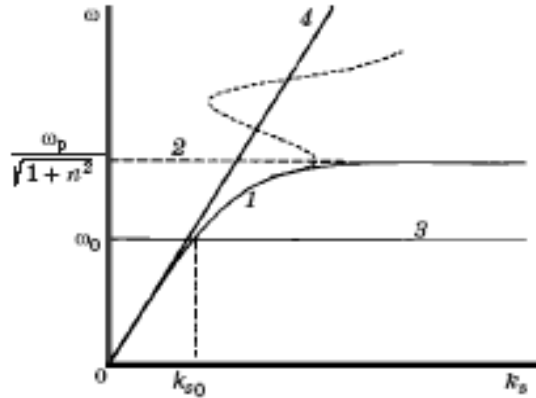


Fig. 1. Typical dispersion curve for surface plasmon polaritons of plane boundary surface-active media – semiconductor with dissipation: 1 – dispersion curve, 2 – asymptotically limited value of dispersion curve for lossless media and $k_s \rightarrow \infty$, n – refractive index of semiconductor, 3 – ω_0 – frequency of laser radiation, 4 – light line, k_{s0} – surface plasmon polariton's wavenumber at frequency of laser radiation. The strokes line shows the dispersion curve behavior for high frequency region and loss plasma

Plasmic frequency is determined as $\omega_p = \left(\frac{4\pi n_e e^2}{m} \right)^{1/2}$; where m – a mass of nonequilibrium electron, e

– electron charge, n_e – a concentration of nonequilibrium electrons. Nonequilibrium concentration, which is corresponded of formula (1), was named critical concentration n_c . Critical concentration n_c is less of value of threshold of destruction of irradiated matter and creation surface nanostructures.

Space inhomogeneity of electron concentration near its critical value may be represented as initial center of transformation of laser irradiation to SPP. In this case resonance condition (1) is transformed to

$$\omega = \omega_s = \frac{\omega_p n_e}{\sqrt{2 + n^2}}. \quad (2)$$

Example of its initial center may be local volume with size $d < \lambda$, where d is diameter of local volume of inhomogeneous concentration, λ – laser wavelength.

The comparison of experimental results with the expression for d in the case $d < \lambda$ shows the coincidence (and taking into account the dependence on the depth of relief (refractive index) modulation with the number of pulses). The experimental range of the $N = \text{Re}\eta$ (where η is permittivity) value changes is $1 < \text{Re}\eta < 1,4$ or all types of materials including metals. The higher values of $\text{Re}\eta$ the slower is the phase speed of SPP.

Formation of structures with the period of d on laser irradiated surfaces of condensed matter may be explained with help of universal polyariton model of lazer-induced destruction of the condensed environments and conditioned interference of laser radiation with excited them by surface plasmons.

As shown in [9, 10], at the femtosecond pulses action on the crystalline semiconductors surface in the transparency region results in the three types of periodic structures formation:

1. Microstructures with $d \leq \lambda$ and orientation $\vec{g} \parallel \vec{E}$;
2. Nanostructures with $d \leq \lambda/2n$ and orientation $\vec{g} \parallel \vec{E}$;
3. Nanostructures with $d \leq \lambda/2n$ and orientation $\vec{g} \perp \vec{E}$, spatially situated along the beam propagation direction $\vec{g} \parallel \vec{k}$, where \vec{k} – the wave vector of light.

The formation first type of structures is connected with nonequilibrium phase change and semiconductor metallization, exciting of surface plasmon polaritons on interface with air (vacuum) and their interference with incident irradiation. At the same time the period of formed structures d is determined by relation $d = \lambda / \text{Re } \eta$. Here η is the normalized complex refractive index of air-plasma interface,

$$\eta = k_s / k_0 \sqrt{\varepsilon / \varepsilon + 1}, \text{ where } \varepsilon \omega \text{ is dielectric permittivity of plasma layer, } \text{Re } \eta \geq 1.$$

The formation of second type nanostructures (small-scale nanostructures) is connected with creation of nonequilibrium plasma layer near air-semiconductor interface.

In this case the excitation of surface plasmon polaritons (SPP) is possible. So their mutual interference and surface plasmon interference with incident laser radiation produces the structures with following periods:

$$d_1 = \lambda / n \text{Re } \eta, \quad d_2 = \lambda / 2n\eta. \quad (3)$$

Special interest excites the formation nanostructures of third type, for that $d = d_2$, but observed orthogonal orientation. Note, that in this case structures localized along the for high intensities of laser radiation the formation of metallized cylindrical plasma channel allows the possibility of cylindrical surface plasmon polariton (CSPP) propagation. Mutual interference of counterpropagating along the laser beam track laser has typical diameter 100–200 nm. According to the model developed in [9, 10], cylindrical surface plasmons polaritons creates the conditions of laser track nanostructuring (Fig. 2).

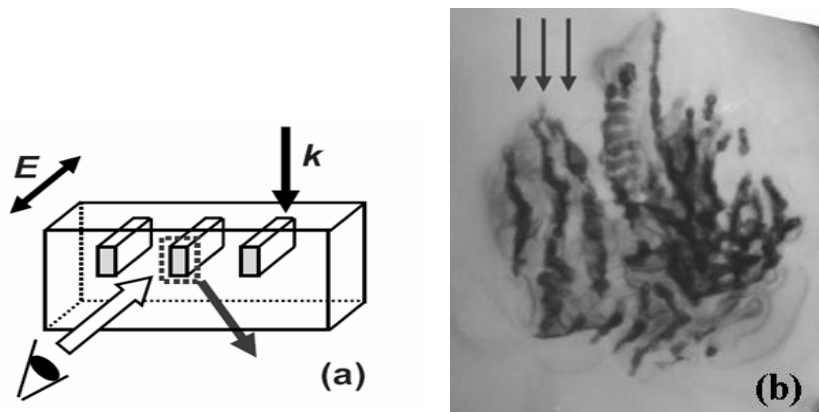


Fig. 2. (a) Schematic illustration of the geometric relationship between the irradiated line and cross-sectional micrograph. (b) Bright field TEM image of the cross section of a line written with pulse energy of 300 nJ/pulse

At the same time it is considered that intensity of radiation is enough for formation in laser pulse the cylindrical plasma channel, that maintain propagation of CSPP (Fig. 2, a). It is clear, that formation of nanostructures occur for series of radiation pulses. Back wave, that go toward the laser radiation and is needed for interference, is created due to reflection by system of formed temporary and remnant regular nanostructures. Since plasma channel is surrounded by semiconductor matter, the period of resulting nanostructure is

$$d_0 = \frac{2\pi}{|2\vec{k}_s|} < \frac{\pi}{k_0 n} = \frac{\lambda}{2\pi} \approx 150 \text{ nm},$$

that is in good agreement with experimental data [9, 10]. The value k_s depends on excess nonequilibrium electrons concentration on the critical one and on refractive index of semiconductor. As for less often observed structures with $d \approx 2d_0 = 300 \text{ nm}$, that their appearance may be connected with nonlinear dynamic of structures in the framework of nonlinear model based on one-dimensional logistic map and Feigenbaum universality [9].

According to the known universal polariton model of destruction surface of the condensed environments, intensity total interference field at influence of the linear polarized laser radiation on a normal to the surface of metal in the conditions of excitation of superficial plasmons it is possible to present in the following kind, taking into account that superficial plasmons spread in direction $\pm x$ (directions of their primary distribution):

$$J_x = I_x + I \cdot I_s^{1/2} \sin g_1 x + \varphi + I_{s_1} \cdot I_{s_2}^{1/2} \sin g_1 x + \psi \quad (4)$$

Here $I(x)$ is intensity of the absorptive laser radiation, $I_{s_i} x$ is depending from a coordinates intensity of absorption of the excited superficial plasmons; indexes of $I = 1, 2$ is conformed to directions of distribution of superficial plasmons in mutually opposite directions of propagation, I_s is total intensity of absorption of superficial plasmons, which is propagated in opposite directions (look also the chart of influence of laser radiation on a Fig. 3, a). Wavevector of basic lattice of $\vec{g}_1 = \vec{k}_{s1} = -\vec{k}_{s2}$, forming of which is conditioned interference of falling radiation and superficial plasmon (see Fig. 3, b), in this case equal to the wavevectors of superficial plasmons, spreading in opposite directions and resulting in forming of double degenerate lattice; wavevector of this lattice $\vec{g}_2 = \vec{k}_{s1} + \vec{k}_{s2}$ is conditioned reciprocal interference of superficial plasmons with opposite directions of propagation (Fig. 3, b); φ, ψ – phase angles between the proper waves. It is assumed in expression (4), that superficial plasmons spread in directions $\pm x$, i.e. in the first approaching ignored the waves of superficial plasmons, spreading in near directions.

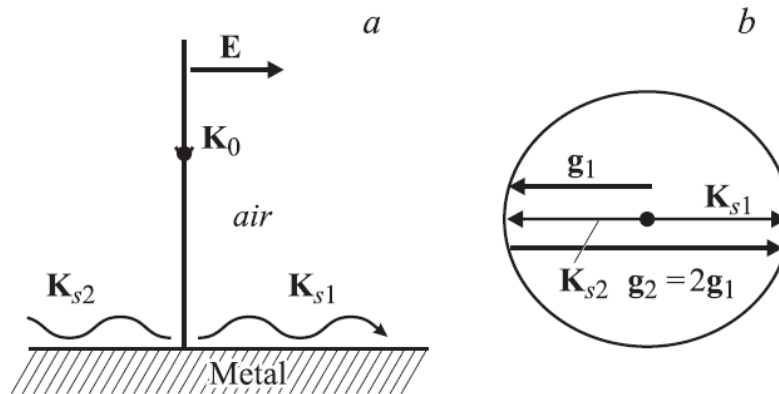


Fig. 3. (a) is a chart of excitation of superficial plasmons k_{s1}, k_{s2} at co-operation of the linear polarized laser radiation, directed on a normal to the surface, with a metal; (b) is the circular vectorial graph, illustrating the conservation law of quasiimpulse and creation of grates of nanorelief on surface of metal due to interference of falling wave with superficial plasmons (grate g_1) and due to mutual interference of superficial plasmons (grate $2g_1$)

First term in (4) gives the permanent constituent I_x in summary intensity of electromagnetic field J_x . At comparatively low intensities of laser radiation (and smalls of heights of grates of resonance nanorelief) a basic contribution to formation of periodic structures gives interference of falling radiation with the superficial plasmons excited them (the second element is in right part of Eq. (4)). Since small values of height of resonance nanorelief (h) size of the electric field of superficial plasmon of $E_s = \xi h E$ [9], on the initial stages of forming of regular surface nanostructure the second term prevails in modulation part of expression (4). Here E is amplitude of the electric field of falling wave, ξ it is a

coefficient of proportion. In these conditions the second term appears proportional $I_s^{1/2} \sim h$. It leads, with the increase of number of impulses of laser radiation N (at the normal falling of radiation), to forming of resonance remaining nanograte of relief with the period of $d = \lambda/\eta$. Here λ is a central wavelength falling

laser radiation, $\eta = \left[\frac{\varepsilon}{\varepsilon + 1} \right]^{1/2}$ is an index of refraction of border of section of metal–vacuum for superficial plasmons, ε – is a permittivity of metal, ω – is central frequency of laser radiation.

At the normal falling of light on the formed grating of g_1 the process of resonance excitation of superficial plasmons, spreading in mutually opposite directions goes simultaneously, with a positive feedback on amplitude of grating of h . With growth of amount of impulses of radiation of N , amplitudes of remaining resonance nanorelief and intensity of the excited superficial plasmons ($I_s > I$) the third term begins to play a basic role in right part of expression (4). This term is represented an interference of superficial plasmons, spreading in mutually opposite directions. Their mutual interference [9, 10], and also interference of the second spatial harmonics of superficial plasmon (wavevector of $k_{s2} = k_{s0} + g_1$, $\omega_i = \omega$, $i=1,2$), with a falling radiation [9, 10, 13] result in forming of degenerate structures with the period of $d = \lambda/2\eta$ and to more effective transformation of energy of falling radiation to the superficial plasmons (SP) (in right part of expression (4) the second term appears small as compared to the third). Here k_{s0} is a wavevector of superficial plasmon for the flat border of section of metal–air, ω_2 is frequency of the second spatial harmonic of superficial plasmon. We will mark that a transition in expression (4) to quadratic dependence on amplitude of relief is possible and in the second term, at large amplitudes of grating of nanorelief [9, 10, 13].

Physical views about the mechanisms of forming of resonance nanostructures (polyariton model) allow to predict forming of nanorelief with spatial periods, multiple $2 \lambda, \lambda/2, \lambda/4, \lambda/8, \dots$, including due to interference of spatial harmonics of superficial plasmons [9, 10, 13]. These periods are corresponded to generation $2n$ -multiply harmonics with point of view of nonlinear optics [12, 13]. It is caused of the second-order nonlinear optical reirradiated effects, which are basic for the formation of proper frozen interference patterns [13].

A mathematical model, explaining forming of structures with multiple periods, is a model of logistic reflection. As a variable quantity of x in a logistic reflection, designing the probed physical process, will examine a root square from rationed on eaten up intensity of falling radiation of size of intensity of absorption of superficial plasmons, $x \sim \left(\frac{I_s}{I} \right)^{1/2} \sim E_s/E$. Intensity of absorption of falling radiation I at multiimpulsive influence increases with growth of N . In the first approaching will suppose, that $J x = \beta N I$ $N=1$. Because of linearness of dependence of the electric field of superficial plasmon of E_s from E at small h [9], $E_s \sim N$. Therefore as a managing (bifurcational) parameter [9] in logistic transformation will enter the size of $\mu \sim \gamma N$. Here β, γ are permanent quantities, not depending from N and amplitudes of the electric field of E . In this case unidimensional logistic reflection, designing the probed physical process, the simple looks like:

$$f(x, \mu) = \mu x(1-x), \quad x \in [0, 1], \quad \mu \in [\mu_1, \mu_2]. \quad (5)$$

It appears that the complex non-periodic conduct of the unidimensional dynamic system is unconnected with originality of logistic transformation [13.5] and takes place for all unimodal transformations, i. e. transformations f , having the unique extremum on the defined interval.

According to Feygenbaum [9], will build bifurcational diagram cascade of doublings of period for the reflection of type (5) (Fig. 4). Here maximum correlation of

$$\lim_{n \rightarrow \infty} \frac{d_{n+1}}{d_n} = \alpha \quad (6)$$

and, determines one of so-called universal Feygenbaum constant and, the exact meaning of which is equal and $\alpha = 2.50290785$ [9, 13]. Universal constant α determines scaling of variable value x in transformation $f(x,y)$. Second variable value μ determines scaling of type

$$\mu_n - \mu_\infty \sim \delta^{-n}. \tag{7}$$

Here $\delta = 4.6692016\dots$ is other universal Feygenbaum constant, μ_∞ is a maximum value of parameter μ_n at $n \rightarrow \infty$.

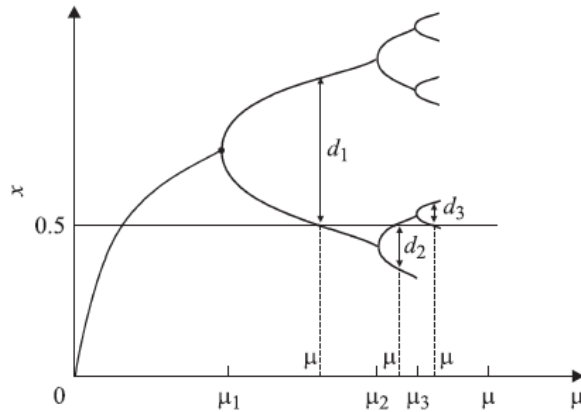


Fig. 4. Bifurcational diagram of reverse Feygenbaum cascade

From the bifurcational diagram of Fig. 4 it is necessary that a size of d_n is distance between the values of $x = 1/2$ and t the nearest to this point elements of circle with period of 2^n $\mu = \mu_n^*$, where μ_n^* is a value of parameter μ , which proper to the supercycle of period of 2^n . This nearest element is 2^{n-1} iteration of point of $x = 1/2$, therefore

$$d_n = f^{2^{n-1}} \left(\frac{1}{2}, \mu_n^* \right) - \frac{1}{2}.$$

Unlike the known models and examples of doubling of Feygenbaum period the reverse doubling of period of dissipative nanostructures, by means of which the nonequilibrium system passes to the complex non-periodic state, will be realized in our case. Unfortunately, the terms of experiment did not allow discovering followings, more small periods of nanostructures. The result of such transition is the expressed fractal character that we continue to name the surface of metal.

Experimental data of doubling of period of diffractive patterns on laser-irradiated materials is corresponded to the interference of waves of second harmonic. Practically we observed this nonlinear effect in frozen state.

We will consider an important question about the dimension (irregular) of attractor of unimodal transformation (5), which gives birth at completion of cascade of Feygenbaum bifurcations (Feygenbaum attractor) [9, 10]. As Feygenbaum attractor is Cantor accomplished nowhere compact set, the use of fractal dimension is most acceptable in this case [238]. Feygenbaum attractor of a point unimodal unidimensional

$$d_f = \frac{\ln 2}{\left[\frac{\ln 2\alpha^2}{\alpha + 1} \right]} = 0,543.$$

transformation is a fractal and has a fractional dimension of

For the case of irradiation the dielectrics by laser with $h\nu > E_g$ the creation of damages in volume in irradiated material has oriental character. This problem is analogous to microscopic mechanism of Cherenkov radiation.

The excitation of the irradiated media may be represented as generation of nonequilibrium electrical dipoles (Fig.5) [7].

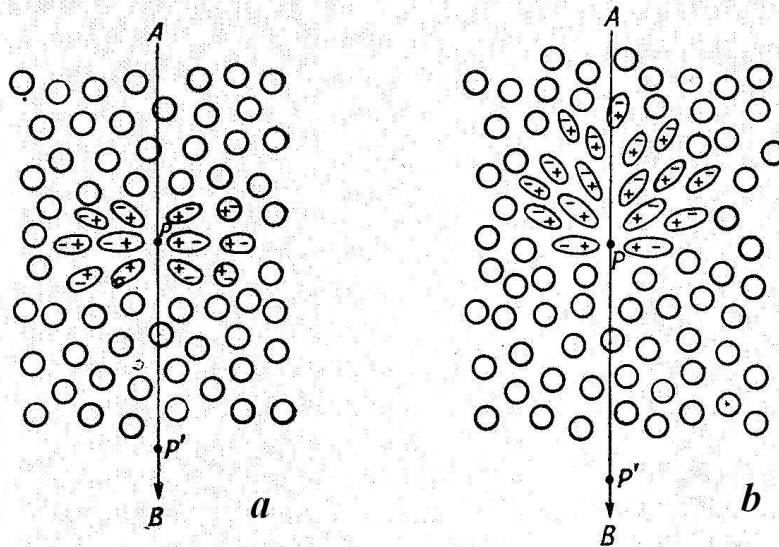


Fig. 5. Polarization, which is generated in dielectric after propagation of charge particle: *a* – with low velocity; *b* – with large velocity

For the low velocity of particle the polarization of generated dipoles in media has random nature (case *a* of Fig. 5). For the large velocity of particle the polarization of generated dipoles in media has ordered nature (case *b* of Fig. 5).

For the laser irradiation these pictures have place for the short intensive regime of irradiation (case *b* of Fig. 5, creation of inverse excitation) and for the more long intensive regime of irradiation (case *a* of Fig. 5, linear or sublinear regime of the irradiation).

These two regimes of irradiation may be having two ways of relaxation: radiant (adiabatic) and nonadiabatic. For adiabatic relaxation of particles excitation we have next types of irradiation: transient radiation for case *a* of Fig. 5 and Cherenkov radiation for case *b* of Fig. 5. For the laser irradiation radiant relaxation is or reirradiation (case *a* of Fig. 5) or lasing generation (case *b* of Fig. 5). Nonadiabatic relaxation is caused the structural changes in irradiated materials. These changes may be having quantum (photoinduced) nature (for the intensive short irradiation of stable structures (crystals) and intensive irradiation of metastable structures (glasses)) and wave nature (thermal, plasmic). The last effects are possible for the intensive regimes of irradiation which are caused high heat, melting and evaporating of irradiated material.

Nonadiabatic relaxation for laser irradiation must be connected with the regime of saturation of excitation. It may be multiphoton absorptive processes. Only in this case we may receive required characteristics of irradiated materials (macroscopic characteristics must be similar to microscopic). Thus, roughly speaking, for nonadiabatic relaxation we can determine cone of creation the damages analogous to Cherenkov radiation with help of next correlation $\theta = \arccos \frac{1}{n}$, where 2θ is angle near vertex of cone, n is refractive index of irradiated material. It allow to explain experimental data of Fig. 2(b). This approximation is true for kinetic and plasmic mechanisms only. For thermal dynamical case it isn't true for all cases.

Conclusions: 1. A problem of creation of laser-induced damages in dielectrics and semiconductors is discussed.

2. Polariton-plasmon model of creation the damages on a surface of irradiated materials is analyzed.

3. Makins Faygenbaum model of nonequilibrium dynamics was used for the explanation of the doubling of period of interference pattern on surface of irradiated materials.

4. Was shown, that methods, which are analogous to methods of microscopic mechanism of Cherenkov radiation, may be used for the explanation of the processes of creation laser-induced damages in volume of irradiated dielectrics on macroscopic level.

5. Represented models are added and expanded the possibilities of Relaxed Optics.

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