Universal Polariton Model of Laser-induced Condensed Matter Damage

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Abstract: The extension of known polariton model of laser-induced condensed matter damage followed by ordered structures formation for the case of ultrashort pulse durations and condensed matter of different physical properties was made. In addition to the usual cases of linear or circular polarizations of laser radiation the case of axi-symmetric polarization was theoretically considered and illustrated by experiments with dielectrics, semiconductors and metals. The special case of radially polarized laser radiation is distinguished as one for which laser-produced dynamic resonant the additional energy of excited surface polaritons into diffraction size focal spot. The advantages of axi-symmetrically polarized laser radiation for materials treatment in framework of universal polariton model were discussed. In experiments the wide variety of spatial periods of microstructures were observed which do not described by existing theories. The nonlinear mathematical model which describes the spatial periods of laser-produced microstructures was suggested. Model describes the formation of microstructures with periods multiplied by laser wavelength and predicts the spatial period's values less than diffraction limit one. The nonlinear theoretical model is illustrated by published experimental data. The idea to explain the cause of the formation of regular nanostructures with anomalous orientation on condensed matter surfaces was suggested. It is based on the effect of wedge (channel) surface plasmon polaritons excitation and their mutual interference under the action of polarized laser radiation. The new phenomenon of laser-induced anisotropy of metal recrystallization under the action of nanosecond duration repetitive pulses of linear polarized radiation was experimentally discovered. The phenomenon was explained as a result of grain-boundary movement by directed flux of skin-layer electrons dragged by surface plasmon polaritons.

1 INTRODUCTION

In recent years many experimental data were published causing ordered micro- and nanostructures formation under interaction of picoseconds and femtosecond polarized laser radiation with condensed media having different physical properties. There is the lack of the models which universally describes such phenomenon in wide range of laser (pulse duration, power density, laser polarization, ...) and material (material properties) parameters. In present article such model is proposed with emphasis on values of spatial period and orientation of formed structures in regimes of ultrashort laser radiation interaction of linear polarization. The extension of polariton model for the case of axi-symmetrical polarization of laser radiation has been made. The variety of spatial ordered micro- and nanostructures produced under the action of laser radiation of long and ultrashort pulses durations observed in many experiments have not been explained in framework of known models. So the problem was exists to create the model which allows an adequate description of periods and orientations of formed structures. The nonlinear mathematical model based on the unimodal logistic map was suggested for this case. In experiments with interaction of pulsed laser radiation tiwh metal it was discovered the anisotropy of recrystallization process governed by the direction of linear polarized laser radiation. In the final stage of laser-metal interaction the quasiperiodic microstructures in the mode of grooves of thermal etching were formed. So the problem arises to elucidate the origin of observed phenomenon. The suggested explanation

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of the phenomenon is based on the drag effect of electrons of metal skin-layer by surface plasmon polaritons (SPP).

2 UNIVERSAL POLARITON MODEL

2.1 Extension of Polariton Model

At first polariton model was suggested for explanation of periodical structures formation under interaction of pulses of laser radiation of long durations (>1 ps), Bonch-Bruevich (1982). The model works in the spectral range of the fulfilment of inequality:

$$\varepsilon_a < -\varepsilon_b \, , \tag{1}$$

for metals, in the conditions of creation of a semiconductor's melt (for semiconductors having semiconductor-metal type phase transition at melting point); for *dielectrics* - in the spectral range of the reststrahlen band (middle IR range). Here $\in_a$, $\in_b$ are dielectric permittivities for the surface active and dielectric media, correspondingly.

The extension of polariton model for *metals* and fs radiation was suggested by Agranat (1999). But the question about applicability of polariton model for *semiconductors* and *dielectrics* was open. Few ideas have been suggested to explain the microstructures production inside and on the dielectrics and semiconductors surfaces. For instance, it was acousto-plasmon model of Shimotsuma (2003), model based on local surface plasmons excitation of Bhardwaj (2006), model accounting for waveguide formation on the semiconductor's surface of Martsinovski (2009).

The main idea suggested by us was the achievement the high concentrations of nonequilibrium curriers in conduction band such as during the pulse semiconductor (dielectric) will have metallic properties. To achieve this, the following inequality must be fulfilled

$$\omega_0 < \omega_{sp0} \, , \tag{2}$$

of excited and usual dielectric, see Fig.1. Each case the limiting frequency of SPPs ω_{sp0} is higher than the nonequilibrium carriers, m and e are mass and charge of electron, correspondingly. The inequality (2) is illustrated with the help of dispersion relation where $\omega_{sp0}=\omega_p/(1+n^2)^{1/2}$, $\omega_p^2=4\pi n_e e^2/m$ where ω_p is the frequency of laser radiation, n is the refractive index of dielectric, n_e is the concentration of laser frequency the possibility of existence and excitation of SPPs exists. Simple estimation shows that critical concentration of nonequilibrium carriers for semiconductor silicon is n_e~2 10^{21} cm^{-3}. This concentration can be easily achieved in experiments with femtosecond laser radiation.

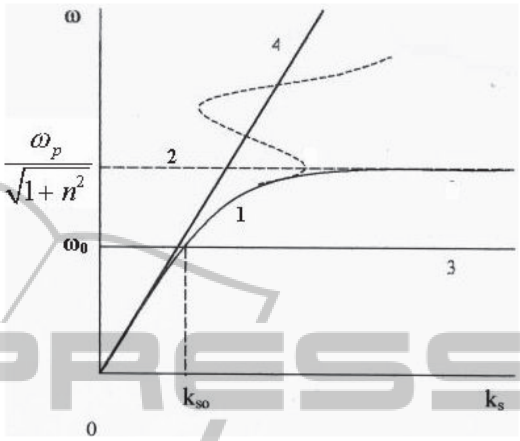

Figure 1: Dispersion curve for plane boundary of surface-active media – semiconductor for surface plasmons with dissipations. 1 – dispersion curve; 2 – asymptotically limiting value of dispersion curve for $k_s \rightarrow \infty$; 3 – line of laser radiation; 4 – light line; k_{s0} – wave number of SPP at laser frequency. Dotted line shows high frequency dispersion curve behavior for medium with loss.

2.2 Spatial Periods of Structures

The periods of spatial structures predicted by old polariton model for long pulse durations (normal incidence of radiation) have two values which originate from interference of incident laser radiation with generated by it SPPs, and mutual interference of SPPs (of opposite propagation directions): d=λ/η and d=λ/2η. Here η is the real part of the refractive index for SPPs of considered boundary. The old model has not explain more wider set of periods arises in experiments, for instance, appearance proportional to λ and multiples by natural numbers periods. Wide variety of periods appeared in femtosecond laser radiation experiments with dielectrics, semiconductors and metals was one of the obstacles to apply the polariton model. So the question arises to explain this variety of periods.

We suggested the nonlinear mathematical model to describe the values of periods of laser-induced structures Makin (2008). The model is founded on basic models from simple nonlinear one-dimensional unimodal logistic maps. We follow the close similarity the expression for total absorbed intensity $\mathcal{J}$ of laser radiation in the case of SPPs excitation:

$$\mathcal{I}(x)=I(x)+(I\cdot I_s)^{1/2}\cos(g_1x+\varphi)+(I_{s1}I_{s2})^{1/2}\cos(g_2x+\psi) \quad (3)$$

and expression for logistic map (4). Here I_s is the intensity of SPPs, I is the intensity of laser radiation, φ, ψ are initial phases, g_1 and g_2 are formed gratings. Let us introduce new variable $x=I_s/I$. We will describe our system by one-dimensional unimodal logistic map:

$$f(\mu,x)=1+\mu x(1-x),\ \ x\in[0,1],\ \ \mu\in[\mu_1,\mu_2]\ . \quad (4)$$

The bifurcation diagram in coordinate (x-μ) which corresponds to logistic map (4) is presented on Fig. 2 and describes the set of bifurcations for definite values of μ_i (i=1, 2, …). Value d_n is the distance between values x=1/2 and nearest to this point element of circle of period 2^n for $\mu=\mu_n^*$, where μ_n^* nearest element is 2^{n-1} iteration of point x=1/2:

$$d_n = f^{2^{n-1}}(\frac{1}{2}\mu_n^*)-\frac{1}{2} \quad (5)$$

μ_n^* is the value of parameter μ, corresponding to supercircle of period 2^n. So the model describes the set of periods (reversed Feigenbaum's cascade) multipled by ½:

$$\ldots,\ 4\lambda/\eta,\ 2\lambda/\eta,\ \lambda/\eta,\ \lambda/2\eta,\ \lambda/4\eta,\lambda/8\eta,\ \ldots \quad (6)$$

The Feigenbaum's universality is the part of Sharkovski order. So the nonlinear mathematical model can be generalized for Sharkovski order.

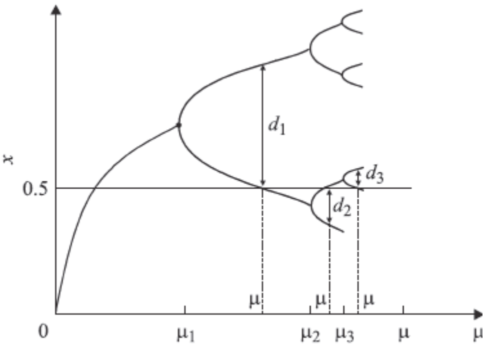

Figure 2: Bifurcation diagram of reverse Feigenbaum's cascade.

Important, that if the period 3 of Sharkovski order is experimentally realized the appearance of any period of Sharkovski order is possible. And this is the rout to chaotic behavior of the system. The obtained results are well illustrated by published experimental results including predicted values of periods far less diffraction limit value Makin (2012). The obtained theoretical results causing the nanostructures formation with periods less than the diffraction limit value are in frame of recently developed nonlinear Abbe theory of Barsi and Fleischer (2013).

2.3 Axi-symmetrical Polarization

Let us consider the action of radially polarized laser radiation (RPR) with selective direction of polarization $\varphi=\varphi_0$ where φ is the angle characterizing (local) direction of the electric field vector of the incident wave. It is known that the SPP frequency is equal to that of the laser radiation, while its wave vector $\mathbf{k}_s$ is determined by the law of conservation of momentum (quasi-momentum):

$$\mathbf{k}_s= \mathbf{k}_t +m\mathbf{g}, \quad (7)$$

where $\mathbf{k}_t$ is the tangent component of the wave vector of laser radiation, **g** is the vector of resonance grating that allows conversion of laser radiation to the surface polaritons (SP), and m is the order of diffraction. At normal incidence of radiation $\mathbf{k}_t=0$, $\mathbf{k}_s=\mathbf{g}$, and

$$\mathbf{g} \parallel \mathbf{k}_s(\varphi_0). \quad (8)$$

So the induced grating is characterized by orientation (8) and period of axial ring structures with a period determined by expression (9), where η is the real part of the refractive index of the interface for SPPs. If $\varphi\in[0, 2\pi]$, i.e., takes all possible values, there appears a grating in the form of circular rings.

$$d=2\pi/|\mathbf{k}_s|=\lambda/\eta, \quad (9)$$

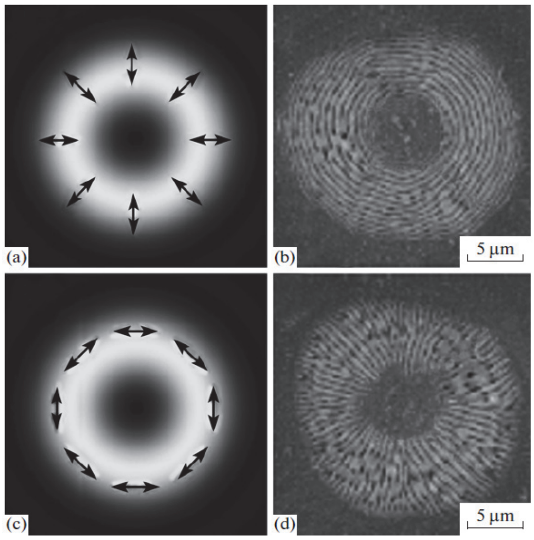

Figure 3: Distribution of (a, c) intensity and (b, d) corresponding microstructures formed under the influence of a train of femtosecond laser pulses with (a) radial and (c) azimuthal polarization on the surface of a silicon, Lou (2012).

Hence, the resonance grating corresponding to the interference of the radially polarized laser radiation with driven by it SPs represents axi-symmetric ring structures with a period determined by (9). The experimental results obtained by Lou (2012) and Hnatovski (2011) support the above consideration; namely, it was shown that radially polarized femtosecond laser radiation interacting with the surface of silicon induced an axisymmertic ring structure with period $d=\lambda/\eta\approx690$ nm and depth up to 300 nm ($\lambda=806$ nm, $\tau=35$ fs, $Q=0.26$ J/cm^2), Fig. 3b.

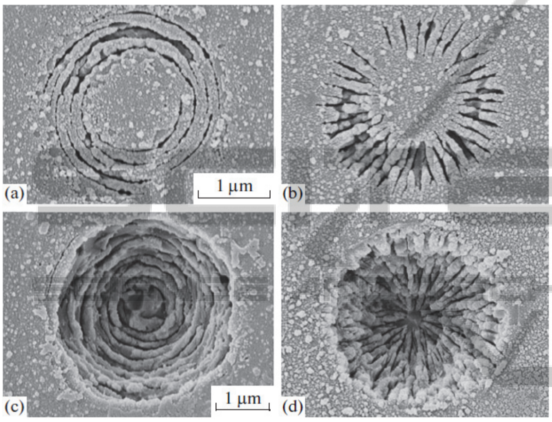

Figure 4: Ordered microstructures formed on the surface of fused silica by a train femtosecond laser pulses ($\lambda=775$ nm, $\tau=200$ fs): (a, c) radially polarized radiation; (b, d) azimuthally polarized radiation. The power densities in Fig. 4c and 4d are 1.6 times higher than in Figs. 4a and 4b, Hnatovski (2011).

A similar structure was observed on the surface of fused silica ($d=\lambda/n\zeta\approx260$ nm, $\lambda=775$ nm, $\tau=200$ fs) Fig 4a. Here, n is the refractive index of fused silica $n\zeta=\eta$. The high power density of the laser radiation used by Lou (2012) and Hnatovski (2011) has induced nonthermal phase transition, as a result of which the medium was acquiring the properties of surface active agent.

Based on similar arguments, it can readily be shown that the azimuthally polarized laser radiation causes formation of radial structures of damage (see Figs 3c, 3d, 4b, 4d). Note, that formation of structures was interpreted by Hnatowski (2011) as a "print" of the field under tight focusing, while no interpretation was offered by Lou (2012).

The interaction of axi-symmetrically polarized laser radiation with metal (stainless steel) also causes the formation of corresponding axisymmetrical gratings Allegre (2013), according to theoretical considerations, see Fig.5.

The character of the formed structures can be explained based on other considerations. It is known that RPR interacting with axisymmetric ring resonance grating excite SPPs propagating in the radial direction, Zhan (2006). The opposite statement is also correct: interference of the RPR with radially propagating SPPs produces an axi-symmetric grating.

If SPPs propagate towards the center of the structure, additional absorption related to their excitaton and energy transfer in the direction of the structure center (Fig. 6) will cause formation of a peak of intensity due to the waves being in phase in the center of the structure. Indeed, in the area where the fields of the laser radiation and the excited SPPs add together, the total field has the form:

$$\mathbf{E}=\mathbf{E}_0+\mathbf{E}_s\,, \tag{10}$$

where

$$\mathbf{E}_s=A(\boldsymbol{z}-\boldsymbol{r}\, i\alpha/k_s)H_1^{(1)}(\mathbf{k}_s\mathbf{r})\exp(i\mathbf{k}_s\mathbf{r})\exp(-i\omega t). \tag{11}$$

A is the amplitude, $\mathbf{r}$ is the radius vector, $\boldsymbol{r}$ and $\boldsymbol{z}$ are the unit vetors, $i\alpha=k_{sz}$, $H_1^{(1)}$ is the first-order Hankel function of first kind, ω is the frequency, and k_s is the SPP wave number. Expression (11) is valid if $k_sr>>1$, i.e., for $r>>\lambda/2\pi$.

The problem of additional power P(r) redistribution due to SPPs excitation and their energy transfer towards the center of irradiated zone of radius r_0 was formulated and solved by Makin (2013). As a result, assuming that $r_0\approx\alpha$, $L=\alpha^{-1}\sim\lambda^2$:

$$P(r)/S|_{r=rd}\approx[1-1/(P_0r_o)]P_0 8L/\pi\lambda^2\, 1/\lambda\sim\lambda^{-1}\,, \tag{12}$$

where $S|_{r=rd}$ is area of SPPs focusing with $r=r_d$, r_d is diffraction limited radius, P_0 is constant, L is the SPP's propagation length along the considered boundary. Hence, the RPR (in the universal polariton model) ensure formation of "hot" areas under the conditions of additional (other than Fresnel) absorption of energy of the laser radiation and its focusing through the SPP energy transfer towards the structure center. The effect of additional absorption of energy of laser radiation by resonance dynamic microrelief gratings, Bazhenov (1986), should take place also in the case of processing of metals by radially polarized radiation of high-power continuous-wave technological CO_2 lasers. The experimentally observed increase in the laser being processed (at the front and the walls) has a processing efficiency by radially polarized radiation by a factor of 1.5 – 2 (data obtained by Trumpf, Niziev (1999)) was attributed to tighter focusing geometry, Dorn (2003), and the fact that the coefficient of absorption of radiation by the surface maximum possible value corresponding to absorption of a p-polarized wave.

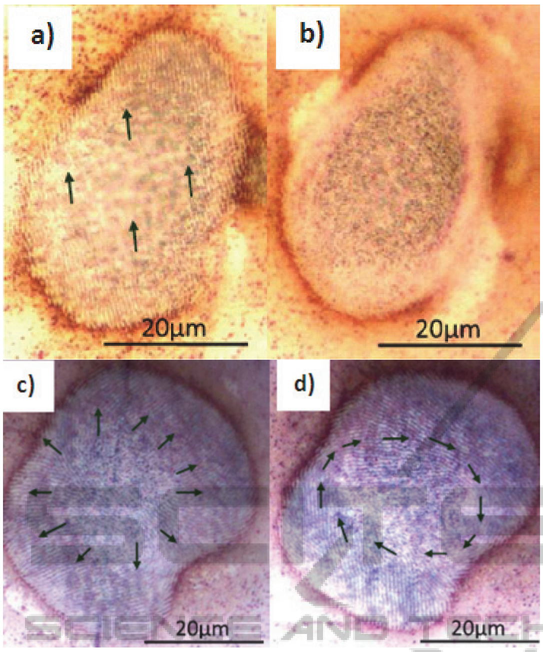

Figure 5: Optical microphotos illustrated polarization vector of laser radiation (λ=800 nm, τ=15 fs, Q=1.5 J/cm^2) distribution and formaion at thefocal plane at stainless steel surface ordered microstructures. The arrows show the direction of the structure **s**⊥**g**, **g** || **E.** Modal convertor (SLM) allows to get beams of laser radiation with linear, ircular, radial and azimuthal polariation and corresponding areas of laser-indued surfae dmage: (a), (b), (c), (d), Allegre (2013).

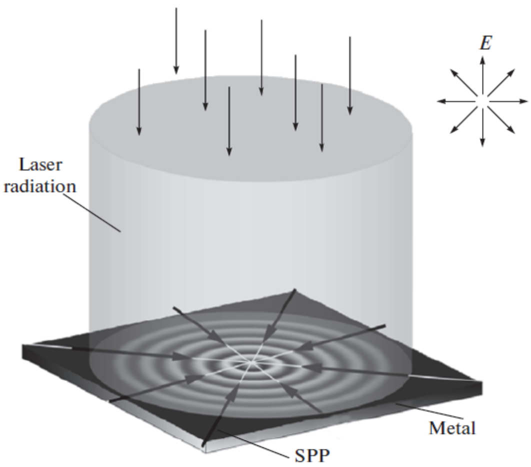

Figure 6: Schematic illustration of formation of an axisymmetric ring resonant grating on the surface of a condensed media by radially polarized laser radiation; the grating converts incident radiation into surface polaritons that are focused onto the structure center. The inset shows polarization of laser radiation in the beam cross section.

The magnification of the electronic image of the surface of silicon (Fig.3b) allowed seeing small-scale structures covering ridges of the main resonance structures of the ring type with period d=230 nm. The grating vector of the small-scale structure was oriented approximately along the tangent line to the ring structures. There structures are the result of excitation of channels SPPs propagating along the ridges of the ring structures of the main resonance relief pattern and their contribution to interference (see section 2.3), Makin (2012).

Ring structures with period $d=\lambda/4\eta=\lambda/4n\zeta\approx125$ nm could also be seen on the surface of fused silica under irradiation with a radially polarized radiation (Fig. 4a); simultaneously, the initial stage of formation of structures with $d=\lambda/8n\zeta\approx64$ nm (upper left corner) could be seen. Here, n(=1.45) is the refractive index of fused silica. The image also reveals nanostructures with d≈40 nm the grating vector of which is parallel to the tangent line to the axi-symmetric grating. The characteristic scale of structures in Fig.4c obtained at higher power densities of laser radiation was also d≈130 nm. The fact that the theoretical expression governing the period of the resonance structures for fused silica contains index n indicates that the structures are localized in glass volume in the near surface layer Makin (2012). Formation of ring-type nanostructures with a period of a multiple of 2 is related to Feigenbaum's universality Makin (2008) (see the section 2.2). Formation of ordered nanostructures with such small periods in glass (k=4, 8) is observed and interpreted for the first time (see, e.g., Makin (2013)).

Axi-symmetric ring-type gratings were experimentally observed in the regime of interaction of long CO_2 laser pulses ($\tau\leq1\mu$s) with fused silica and were caused by interference of unpolarized radiation with surface phonon polaritons driven by it.

Note that the interpreted effect of formation of ordered damage structures by nontraditionally polarized radiation can be used for creation of devices controlling the polarization of laser radiation via reconstruction of polarization from the character of spatial distribution of produced ordered structures (solution of the inverse problem).

2.4 Anomalous Grating Orientation

In the interaction of polarized femtosecond laser radiation the formation of nanostructures with anomalous orientation **g**⊥**E**$_t$ (normal incidence) was observed for condensed matter with physically different properties: metals, semiconductors, dielectrics by Makin (2009). The period d of such structures was small in comparison with wavelength of incident radiation, usually d<<λ. Recently we have observed the formation of such structures in regime of long pulse durations (titanium, germanium Makin (2011)). The origin of their appearance was not clear. We proposed the model based on the mutual interference of so called wedge (or channel) surface plasmon polaritons propagating along the ridges (hollows) of main resonant structures (with opposite propagation directions). The dispersion relation of such wedge (channel) SPPs can sufficiently differ from one for planar boundary (their dispersion curve lies under the curve for plane boundary SPPs curve). So the periods of structures which corresponds to such interference can be as small as ~40-60 nm for metals, ~50-60 nm for compound semiconductors and ~40 nm for quartz glass and corresponding gratings have anomalous orientation (see section 2.2). Note that excitation of wedge (channel) SPPs is one more way for laser radiation energy transfer into heat dissipated inside irradiated materials.

2.5 Quasi-periodic Gratings on Metal

In our experiments the effect of formation of quasiperiodic gratings on surface of polished titanium under the action of 10 ns pulses of linear polarized laser radiation (λ=1,06 μm, τ=10 ns) was discovered. The process of anisotropic metal recrystallization governed by polarization of laser radiation was experimentally observed. The period of produced structures in mode of grooves of thermal etching having orientation $\mathbf{g}\perp\mathbf{E}_t$ was ≈5 μm and rises to 6 μm with laser power density. Later analogous experimental data were achieved for titanium and amorphous titanium alloys for femtosecond laser pulse durations. To explain the observed phenomenon the physical model was suggested by Makin (2013). The model involves the excitation of SPPs by the grain boundaries in the preferential direction of electric field vector of incident laser radiation, drags of electrons of skin-layer by SPPs and movement of grain boundaries, step by step from pulse to pulse, via transfer of electron's momentum to them.

3 CONCLUSIONS

The extension of polariton model for the case of ultrashort laser pulse durations and materials with different physical properties: metals, semiconductors, and dielectrics was made. The nonlinear mathematical model to describe the spatial periods of laser-induced structures in the process of laser-matter interaction involving excitation of surface polaritons was suggested and experimentally verified for different materials: dielectrics, metals, ceramics and semiconductors. The nonlinear model describes the formation of structures with periods proportional to λ and sufficiently lower values of periods in comparison with diffraction limit value. The resonant axi-symmetrical micro- and nanostructures structures which arise under the interaction of axi-symmetric polarization of laser radiation with matter were theoretically predicted and experimentally illustrated for metals, semiconductors and dielectrics.

The origin of formation of the nanostructures with anomalous orientation $\mathbf{g}\perp\mathbf{E}_t$ for long and ultrashort laser pulse durations were cleared as a result of excited wedge or channel SPPs participation in interference process.

The effect of anisotropy of metal recrystallization under the action of long and ultrashort laser pulse durations of linear polarized laser radiation was discovered. The theoretical explanation of observed effect was suggested based on the effect of grain boundary movement by electrons of skin-layer dragged by surface plasmon polaritons.

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