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Problems of modeling the surface interference relaxed optical processes and phenomena

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Problems of created and modeling of surface interference relaxed optical processes and phenomena are analyzed. Problem of nonlinear fundamental absorption and its role in the formation of surface irreversible interference pattern is discussed. Comparative analyses of dynamical (plasmic and thermal) and kinetic (physical-chemical) models is represented too.
Keywords: interference, Relaxed Optics, irreversible processes, fundamental absorption, Nonlinear Optics.

Проаналізовано проблеми створення та моделювання поверхневих інтерференційних релаксаційнооптичних процесів та явищ. Обговорюється проблема нелінійного власного поглинання та її роль у формуванні поверхневих незворотних інтерфереограм. Також проведено системний аналіз динамічних (плазмових і теплових) та кінетичних (фізико-хімічних) теорій і моделей. Акцентується увага на плазмовій (електродинамічній) природі утворення поверхневих інтерференційних структур. Показано, що фізико-хімічна каскадна модель дозволяє пояснити мікроскопічні механізми особливості утворення поверхневих наноструктур. Висловлюється та обґрунтовується припущення про розширення нелінійнооптичних явищ на незворотні (релаксаційнооптичні) процеси: багатофотонне поглинання призводить до фазових трансформацій опроміненого матеріалу.
Ключові слова: інтерференція, релаксаційна оптика, незворотні процеси, власне поглинання, нелінійна оптика.

Проанализированы проблемы создания и моделирования поверхностных интерференционных релаксационнооптических процессов и явлений. Обсуждается проблема нелинейного собственного поглощения и ее роль в формировании поверхностных необратимых интерфереограм. Также, проведен системный анализ динамических (плазменных и тепловых) и кинетических (физико-химических) теорий и моделей. Акцентируется внимание на плазменной (электродинамической) природе образования поверхностных интерференционных структур. Показано, что физико-химическая каскадная модель позволяет объяснить микроскопические механизмы и особенности образования поверхностных наноструктур. Высказывается и обосновывается предположение о расширении нелинейно оптических явлений на необратимые (релаксацийнооптические) процессы: многофотонное поглощение приводит к фазовым трансформациям облученного материала.
Ключевые слова: интерференция, релаксационная оптика, необратимые процессы, собственное поглощение, нелинейная оптика.

Inroduction

Problem of modeling irreversible laser-induced nanostructures is one of central problems of Relaxed Optics (RO) [1, 2]. This problem is connected with electrodynamical processes of redistribution first-order and second wave processes in irradiated materials.

First-order processes are the excitation and ionization of proper centers of light scattering and chemical bonds [1, 2]. Second-order processes are the processes of the relaxation and interaction of first-order excited states.

Laser-induced interference processes and phenomena are the second-order processes. These processes have two aspects: electrodynamical (optical) and physical-chemical. First part is corresponded to redistribution of results of interaction of electromagnetic oscillations, which cause the creation of interferogramms on surface of laser irradiated matter. These processes and phenomena may be explained with help of theories thermodynamical creation nanostructures [3] and surface polariton-plasmon [4]. Second part is corresponded to creation of polyphasic structure in each nanohill [3] or nanocolumn [4], including the change of stoichiometry of irradiated materials. These phenomena must be observed with help of physical chemical methods and methods of phase transformations.

Results and discussions

First irreversible laser-induced interferograms were received by M. Birnbaum in 1965 after pulse Ruby laser irradiation of germanium [5].

Results of more detail research of laser-induced interferograms ion semiconductors were received by I. A. Sokolov [6].

Periodic laser-induced surface structures were received an amplified *Ti* with help sapphire laser system

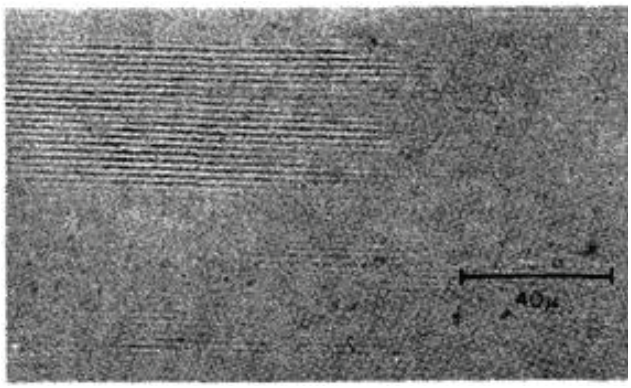

Fig.1. Photomicrograph of Ruby laser induced surface damage of {100} face of a germanium sample [5].

that generates 65 *fs* pulses with energy around 1,1 *mJ*/ pulse at a maximum repetition rate of 1 *kHz* and with a central wavelength λ=800 nm [4]. The laser beam was horizontally polarized and was focused normally onto in metal sample that is vertically mounted on an X-Y motorized translation stage. Scanning electron microscopy (SEM) images of femtosecond laser-induced periodic surface structures (FLIPSS) are represented in Fig. 2. These

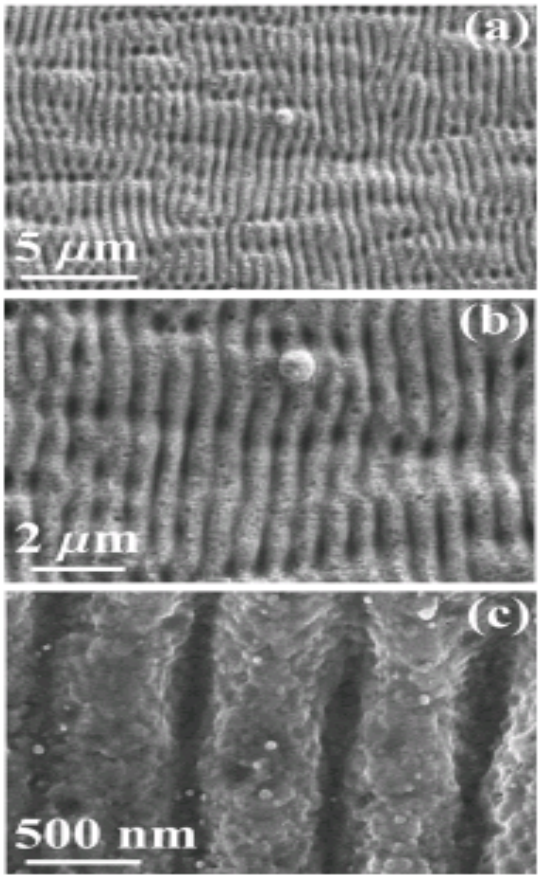

Fig. 2. SEM images of FLIPSSs on silver produced by a scanning femtosecond laser beam. [(a) and (b)] Microscale futures of FLIPSSs. (c) Nanoscale futures of FLIPSSs [4].

FLIPSSs were produced using a scanning laser beam with fluence of 0,16 J/cm^2. FLIPSS period in Fig. 2 is measured 575–625 *nm* [376] and this value is significantly less then the laser wavelength (800 *nm*). Nature of these interferograms are caused of generation of surface plasmons [4].

Further researches shown that these interferograms have nonostructural nature (nanohills and nanocolumns) [1-4].

AFM 3D image of *GaAs* surface after irradiation by YAG:Nd laser at density of power I=5.5 MW/cm^2 is represented on Fig. 3 [3]. Nanohills have various high and place in the maximums of interferograms.

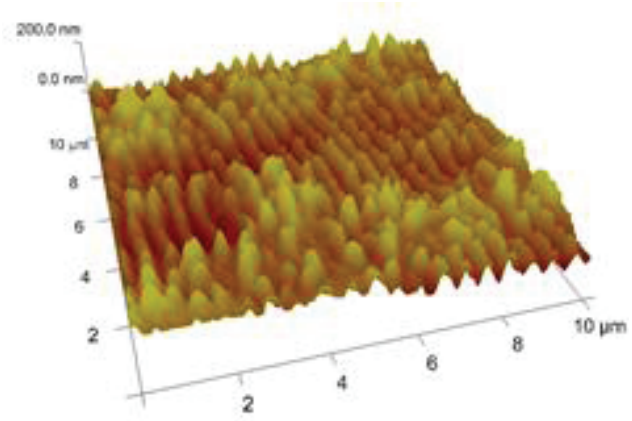

Fig.3. AFM 3D image of GaAs surface after irradiation by second harmonic Nd:YAG laser at I=5.5 MW/cm^2 [3].

Analogous nanostructures were received after irradiation of SiO_2/Si structure by second harmonic Nd:YAG laser at density of power I=2.0 MW/cm^2, pulse duration 10 *ns*, wavelength 532 *nm* and frequency of repetition 12,5 *Hz* (Fig.4) [3].

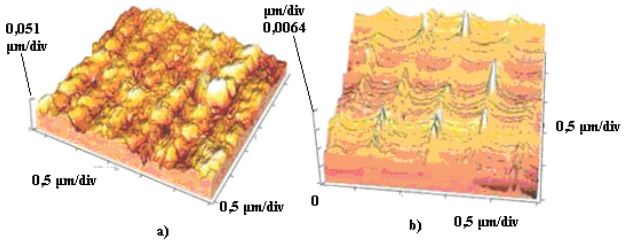

Fig. 4. AFM 3D images of: (a) SiO_2 surface after irradiation of SiO_2/Si structure by second harmonic Nd:YAG laser at I=2.0 MW/cm^2 and (b) *Si* surface after subsequent removing of SiO_2 by *HF* acid. [3].

The results of increasing of height of nanocones to 100 nm were received on Ge after second harmonic Nd-laser irradiation with power density 28 MW/cm^2. The 3D picture of the irradiated surface of a Ge samples as seen under AFM is shown in Fig. 5 [3].

Height, form, geometry and physical and chemical structure of nanohills is depended from conditions of irradiation.

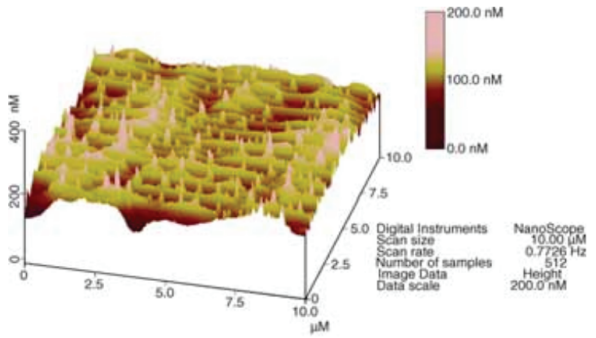

Fig. 5. Three-dimensional AFM image of self-organized nanostructures Nd:YAG laser radiation at intensity 28 MW/cm^2 [3].

Ordered structures, which were created on surface of Si after laser irradiation through lay of water, are represented on Fig. 6 [4]. Three types of micro and nanostructures are generated. Nanostructures have typical spatial scale d_1=600 *nm* and d_2=120 *nm*, here lattice vector oriented $\vec{g} \parallel \vec{E}$. It is corresponded to interference between surface polariton-plasmon (SPP) and TM electromagnetic wave. Structures with period d_1 are generated for interference of falling wave with SPP wave, which arise on the border water – free electrons of silicon. Structures with period d_2 are generated for interference mutual interference of two SPP, which were propagated in mutually inverse directions along border silicon – plasmic layer. Structures with period 120 *nm* aren't depended from nature of liquid, which was contacted with silicon. It is experimental fact.

Third type of nanostructures (d_3=90 *nm*) was generated

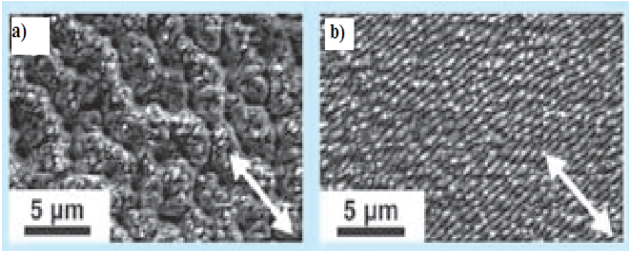

Fig. 6. Ordered structures, which were generated on surface of silicon after laser irradiation through lay of water, (arrow in lower angle show the direction of polarization of laser radiation); duration of pulse 100 *fs*, wavelength – 800 *nm*, number of pulses 200, density of energy the irradiation a) 25 *kJ*/*m*^{2}, b) 5 *kJ*/*m*^{2} [4].

after irradiation structures with d_2=120 *nm* after irradiation of changing polarization, when orientation of vector $\vec{E}$ was changed on 90° relatively to initial action. Power of laser irradiation was less in two time as for previous case. Generated periodical structures (Fig.7 and Fig. 8) are nanocolumns with height to 400 *nm* with spatial period 90 *nm* and orientation $\vec{g} \parallel \vec{E}$.

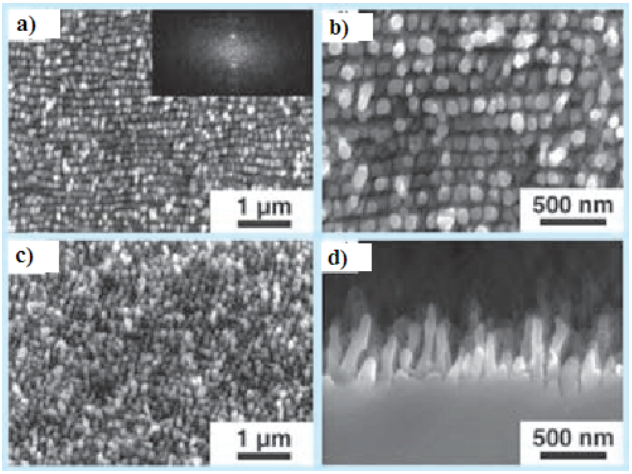

Fig. 7. Nanocolumns, which are generated after irradiation structures of Fig. 6, (wavelength of irradiation 800 *nm*, number of pulses – 200, density of energy of irradiation 0,5 *kJ*/*m*^{2}): a) and b) turn of polarization on 90°, c) turn of polarization on 45°, d) cross chip of nanocolumns. On insertion to Fig. 14a – Fourier-picture of structures [4].

Axes of nanocolumns have perpendicular orientation to initial surface. Structures on other crests are differed slightly in periods and weren't correlated in phase (Fig.4.15a). Moreover Fourier transformation of these structures confirms here periodicity in direction of lattice vector $\vec{g}$ of initial nanostructures (Fig.6 and Fig. 7).

Generation of periodical nanostructures along crests (d=90 nm) is cause with interference of falling radiation with SP, which are exited along crest of relief (d~120 *nm*), and with mutual interference of SPP. A crest of relief, which

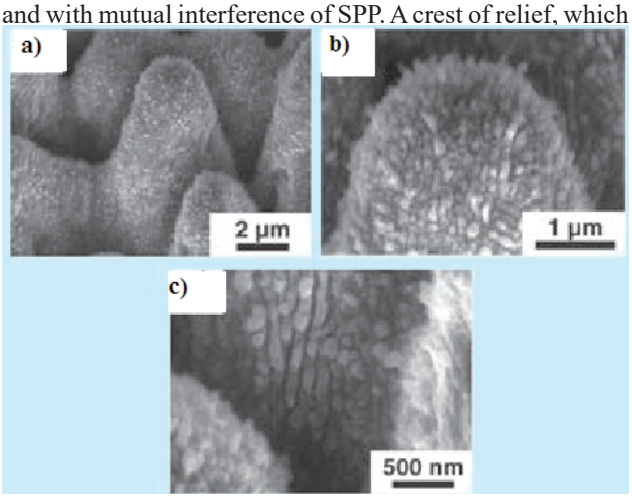

Fig. 8. Surface nanocolumns of little scale, which have orthogonal orientation to a crests of nanorelief of large scale [4].

considered in contact with the substrate, was selected as initial half-cylinder. Formed in this case inoculating regular relief d~90 *nm* is basis for further growth of nanocolumns. Since typical radius of half-cylinder $r < \lambda$, therefore

dispersion relation for SPP in cylindrical geometry is changed from dispersion relation in plane geometry of phase separation. It cause to formation nanostructures with less period as for plane case.

For case of elliptic polarization and falling angle to surface from 0° to 20° basic nanostructures are created: 1) surface nanostructures with period ~200 *nm* and 2) these structure with period 70–100 *nm* are generated on crest of structure 1, but its orientation $\vec{g} \perp \vec{E}$ [4].

Phenomena of doubling of period of laser-induced surface structures is represented in Fig. 9.

Data of Fig. 9 were explained with help help nonlinear Feygenbaum dynamics [4]. Roughly speaking this phenomenon may be represented as example of «structural» generation of second harmonic. According to I. R. Shen this phenomenon isn't be observed for self-absorption range [7]. In classic Nonlinear Optics it is impossible, but in Relaxed Optics it is possible [1,2]. Radiated relaxed processes of optical absorption in matter are caused Linear and Nonlinear Optical phenomena and radiationless relaxation – Relaxed Optical phenomena (phase transformations of irradiated matter).

Universal polariton surface concept may be explained on the basis of Fig. 10.

According to the known universal polyariton 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

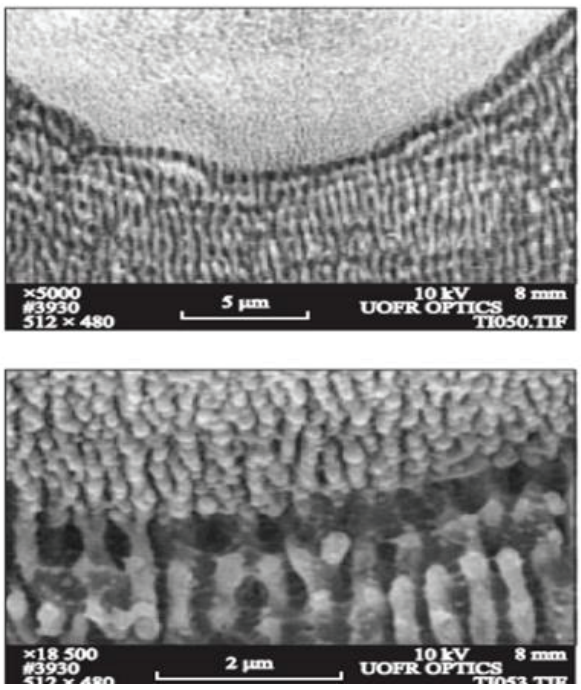

Fig. 9. Relief of Ti surface after pulse linear polarized laser irradiation (power density 1,1 GW/cm^2, number of pulses – 100): period of structures – 600 nm, b) region of transition from structures with priod 600 nm to period 300 nm.

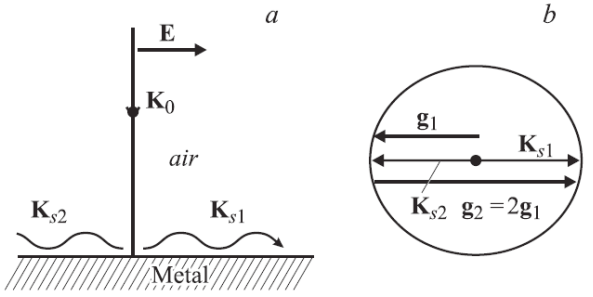

Fig. 10. a – is a chart of excitation of superficial plasmons (k_{s1}, k_{s2}) at co-operation of the linear polarized laser radiation, directed on a normal to the surface, with a metal; *b* – is the circular vectorial graph, illustrating the conservation law of quasimomentum and creation of grates of 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 $2g_1$).

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 ±*x* (directions of their primary distribution):

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

Where *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. 10*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. 10*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. 3.8*b*); φ, ψ – phase angles between the proper waves. It is assumed in Eq. (1), that superficial plasmons spread in directions ±*x*, i.e. in the first approaching ignored the waves of superficial plasmons, spreading in near directions.

First term in (1) 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. (3.64). Since small values of height of resonance nanorelief (h) size of the electric field of superficial plasmon of $E_s = \xi hE,$ on the initial stages of forming of regular surface nanostructure the second term prevails in modulation part of Eq. (1) [4]. Where 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 pulses of laser radiation N (at the normal falling of radiation), to forming of resonance remaining nanograte of relief with the period of $d = \lambda/\eta$. Were λ is a central wavelength falling laser radiation, $\eta = \left[\varepsilon/\varepsilon + 1\right]^{1/2}$ is an index of refraction of border of section of metal–vacuum for superficial plasmons, $\varepsilon(\omega)$ is a permittivity of metal, ω – is central frequency of laser radiation.

At the normal falling of light on the formed grate of g_1 the process of resonance excitation of superficial plasmons, spreading in mutually opposite directions goes simultaneously, with a positive feed-back on amplitude of grate of h. With growth of amount of pulses 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 formula (1). This term is represented an interference of superficial plasmons, spreading in mutually opposite directions. Their mutual interference [4], and also interference of the second spatial harmonics of superficial plasmon (wavevector of $k_{s2} = k_{s0} + g_1$, $\omega_1 = \omega$, $s = 1,2$), with a falling radiation [4] 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 Eq. (1) 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 Eq. (1) to quadratic dependence on amplitude of relief is possible and in the second term, at large amplitudes of grate of nanorelief [4].

Physical-chemical nature of laser-induced phase transformations in irradiated materials may be observed with help cascade model of step-by-step excitation of chemical bonds in irradiated matter [1, 2].

Straight method of the estimation the energetic characteristics this processes may be realized in the next way. Energy of "disruption" of chemical bonds of one type is equalled

$$\mathrm{E}_{di} = N_i E_i, \qquad (2)$$

where N_i – a density of proper bonds; E_i – energy of a disruption (ionization) one bond.

Density of bonds was determined with help Eq. (3)

$$N_i = \frac{\rho N_A}{CA}, \qquad (3)$$

where ρ – density of semiconductor, N_A – Avogadro number, A – a weight of one gram-atom, C – coordination number.

For $C = 8$ we have $N_{iSi} = 6,26 \cdot 10^{21} cm^{-3},$ and for $N_{iGe} = 5,68 \cdot 10^{21} cm^{-3}$. This method was used for the modelling of laser-induced phase transformations in silicon, germanium and allotropic phases of carbon [9]. It allow to explain basic peculiarities of creation new laser-generated phases in irrradiated materials. It may be used for the pecosecond and femtosecond regimes of irradiation.

Phase diagram of silicon (Fig. 11) was selected for the modelling [10]. Roughly speaking, basic causes of laser-induced generation of interferograms and nanohills are creation of surplus of negative charge and, as result, plasmonic oscillations in subsurface region. Surplus of negative charge is caused symmetry and stehiometry of each nanohill or nanocolumn. For case of binary semiconductors surface and peak of nanohill are rich of acceptor component. Symmetry of each nanohill is decreased from basis to peak. For silicon it may be next chain: structure with CN=8 – structure with CN=6 – structure with CN=5 – structure with CN=3 and quasicrystal modifications [2]. But this scenario is characterized plasmic regime of irradiation, when processes of melting, evaporation and sublimation are negligible. Including thermal characteristics is caused the decreasing and spreading of nanohills. Hear chemical and structural characteristics may be changed too. Therefore for more long-term regimes of irradiation we must allow radiated and thermal relaxation [9].

Basic peculiarities of creation the laser-induced surface nanostructures are next: heght of the proper nanohills (for nanosecond regime of laser irradiation) and nanocolumns (for femtosecond regime of laser irradiation) is depended from intensity and time of irradiation. For nanosecond regime of laser irradiation high of nanohills is equaled 15 – 100 nm [3], for femtosecond regime of laser irradiation height of nanocolumns is equaled 400 – 450 nm [4]. Structure of nanohills for germanium is change from diamond to hexagonal (peaks of nanohills) [3]. Femtosecond nanocolumns must have the chain of crystal structures, which is changed from diamond in the

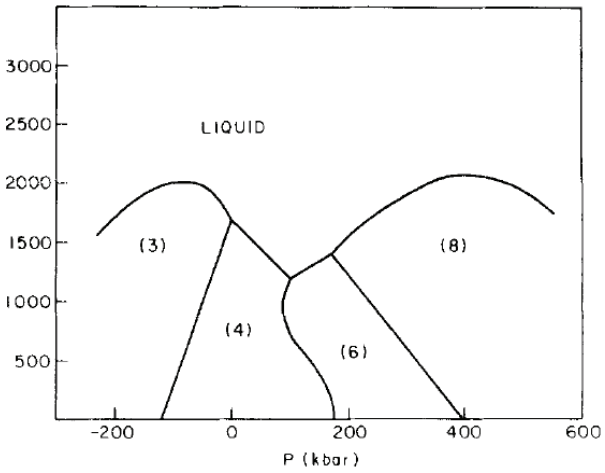

Fig. 11. A schematic phase diagram for *Si*(CN). The coordination numbers (CN) of the various phases are indicated. The diagram is based on common features of the phase diagrams of column IV elements as described by the references cited in Pistorius's review (Ref. 8 in [259]). Starting from a high tempera-ture $>3\times10^3$K and subject to a constraint of average density $\langle\rho\rangle=\rho(4)$, a hot micronucleus will tend to bifurcate into the most stable phases (highest T_m) which straddle *Si*(4) in density. These are *Si*(3) and *Si*(8), as indicated by the diagram [10].

basis nanocolumns to hexagonal or trigonal in the length of proper nanocolumn to direction to its peak. In whole 12 crystal and quasicrystal structures for silicon may be created on the length of proper nanocolumns. Basic limitation of this process is the minimal size of creation of proper phase and thermal processes of relaxation. Each phase structure (nanostructures) has minimal size. This condition is one of basic for the formation of nanostructural pictures of our interferogramms.

With point view of phase transformations the problem of decreasing of crystal symmetry is caused with problem of the creation proper phason – nucleus of new phase [1,2]. Significant parameters of multipulse regime of irradiation are the intensity in one pulse and frequency of pulse repetition. Last must be more as relaxation time of previous excited "phason" state. In this case we have kinetic growth of proper nanocrystall structure. Therefore nanohills of "hexagonal" Ge (Fig. 5) have more large height as for GaAs (Fig. 3) and Si (Fig. 4).

Processes of creation of surface laser-induced nanostructures have electromagnetic nature. This concept is verified of hedgehog-simple surface of first-order nanocolumns after second-order additional irradiation: needles of these hedgehogs are perpendicular to initial surface (Fig. 8).

Therefore for modeling of these processes we must used methods of nonlinear dynamics, including electrodynamics, irreversible physics, physics of phase transformations and methods of quantum chemistry.

Perspective method of modeling may be concept of coherent structures [11]. All possible laser induced processes may be represented as creation and evolution of coherent structures [11].

These results may be represented as expansion of researches of nonlinear and irreversible processes in the self-absorption range of irradiated matter too [1,2].

Conclusions

Comparative analysis of various laser-induced surface nanostructures is represented. Universal polariton and cascade models are represented and used for the explanation basic peculiarities of these processes and phenomena.

1. Trokhimchuck P.P. Relaxed Optics: Necessity of Creation and Problems of Development.// International Journal of Advanced Research in Physical Science, Vol. 2, Is. 3, 2015. – P.22-33.
2. Trokhimchuck P.P. Relaxed Optics: Reality and Perspectives (Review). // Advances in Applied Physics, vol. 3, No.4, 2015. – P. 325 – 341. (In Russian).
3. Medvid' A. Nano-cones Formed on a Surface of Semiconductors by Laser Radiation: Technology Model and Properties. Nanowires Science and Technology, ed. Nicoletta Lupu. – Vukovar: Inech, 2010. – P. 61–82.
4. Makin V.S. Regularities of creation of ordering micro- and nanostructures in condensed matter for laser excitation of mode of surface polaritons. D. Sc.Thesis. – Saint-Petersburg: State University of Informative Technologies, Mechanics and Optics. – 384 p. (In Russian).
5. Birnbaum M. Semiconductor surface damage produced by Ruby Laser.// Journal of Applied Physics, vol. 36, Issue 11, 1965. – P. 3688–3689.
6. Sokolov I.A. Interference laser annealing of semiconductors. Ph.D. Thesis. – Lenigrad: A. Joffe Physical Technical Institute, 1983. – 140 p. (In Russian).
7. Shen Y.R. Principles of nonlinear optics. – Moscow: Nauka, 1989. – 559 p. (In Russian).
8. Landau L. D., Lifshits E. M. Electrodynamics of Condenced Matter. – Moscow: Nauka, 1982. = 621 c. (In Russian).
9. Trokhimchuck P. P. Foundation of Relaxed Optics. Lutsk: Vezha, 2006. – 294 p.; Foundations of Relaxed Optics. Lutsk: Vezha, 2011. – 627 p.
10. Philips J.C. Metastable honeycomb model of laser annealing.//Journal of Applied Physics, No.12, Vol. 52, 1981. – P.7397–7402.
11. Trokhimchuck P.P., Dmytruk I.P. Problem of coherence in modern theoretical physics. //Bulletin of Lesya Ukrayinka East European National University. Physics. No. 2(251), 2013. – P. 46-53.