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Problems of Reradiation and Reabsorption in Nonlinear and Relaxed Optics

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Abstract: *Problems of reemission and reabsorption in Nonlinear and Relaxed Optics are discussed. It was shown that these processes and corresponding phenomena are connected with processes of first order absorptive excitations in irradiated matter and second order relaxation of these excitations. Nature of this relaxation stipulated Nonlinear Optical phenomena for case of radiated relaxation and Relaxed Optical phenomena for case of radiationless relaxation. Basic peculiarities of these processes and methods of its modeling are analyzed and discussed.*

Keywords: *Nonlinear Optics, Relaxed Optics, Pointing tensor, reemission, reabsorption, reirradiation, laser implantation, irreversible phenomena, nonequilibrium phenomena.*

1. INTRODUCTION

Nonlinear optics (NLO) is the branch of optics that describes the behavior of light in nonlinear media, that is, media in which the dielectric polarization P responds nonlinearly to the electric field E of the light. Notion of Nonlinear Optics was introduced from wave differential equation: perturbation of matter may be represented with help nonlinear wave equations [1 – 3]. This nonlinearity is typically only observed at very high light intensities (values of the electric field comparable to interatomic electric fields, typically 10^{10} V/cm) such as those provided by pulsed lasers. Above the Schwinger limit, the vacuum itself is expected to become nonlinear. In nonlinear optics, the superposition principle no longer holds. NLO is researched the behavior and change of radiation in irradiated matter.

Relaxed Optics (RO) is the chapter of modern physics of irreversible interaction light and matter [2, 4, 5]. Necessity of creation RO is caused of technological applications of laser radiation (laser annealing, laser implantation and other [4,5]). Phenomenological energy-time classification of processes and phenomena is basis of RO. According to this classification we have three types of processes and phenomena: kinetic (mainly quantum first-order processes); dynamic (mainly wave second-order processes) and mixing kinetic-dynamic or dynamic-kinetic processes. Roughly speaking RO is the synthesis Quantum Electronics, Nonlinear Optics, Physical Chemistry, Radiation Physics of Status Solid, Physics of Irreversible Phenomena in one system. RO is researched the behavior and phase transformations of irradiated matter.

Problems of reradiation and reabsorption in Nonlinear and Relaxed Optics are one of basic [2,4,5]. Its determine nature of corresponding processes and phenomena. But processes of reemission and reabsorption are common for NLO and RO. Nature of Nonlinear Optical and Relaxed Optical processes is determined of mechanisms of second order (final) relaxation of first order excitation in irradiated matter. Processes, which are researched in NLO, have basically radiative relaxation, whereas processes of RO have basically radiationless relaxation.

But pure Nonlinear or Relaxed Optical processes are occasional phenomena. The problem of interaction of Nonlinear and Relaxed Optical processes is based on the correlation between processes of radiated and non-radiated relaxation of first order excitation (light scattering) and its interference. Roughly speaking, NLO phenomena may be induced irreversible changes in irradiated matter, RO phenomena may be source of NLO effects. These processes are interconnected and mutually complementary phenomena [2,4,5].

2. PROBLEMS OF RERADIATION AND REABSORPTION IN NONLINEAR OPTICS

Processes of multiphotonic absorption and reemission are the basic in NLO. But basic NLO phenomena are realized in intrinsic range of spectrum of irradiated matter. Concentration of optical active impurities is $10^{15} – 10^{17}$ cm^{-3}. Therefore in the case of crystal matter (concentration of knots of crystal lattice is $10^{21} – 10^{23}$ cm^{-3}) we can use the methods of Classic and Quantum Electrodynamics and perturbation theory [1 – 3].

But in NLO we researched the behavior and transformation of optical radiation after interaction first-order radiation with matter. Basic NLO phenomena have reemission nature. The irradiated matter must be stable and can't change in macroscopic sense in the process of irradiation. The nonequilibrium change of irradiated matter is represented with help coefficients of nonlinear susceptibility. As rule these nonlinear coefficients are smaller as permittivity of irradiated matter and it depend from intensity of irradiation [1 – 3].

Classic NLO phenomena are observed in dielectrics. Therefore the permeability of matter $\mu = 1$. These phenomena may be described with help of permittivity ε and its expansion into a series by step electrical strength. For the anisotropic matter permittivity ε must be change on tensor of permittivity ε_{ij}. Expansion into series this tensor has next form [1 – 3]:

$$\varepsilon_{ij} = \varepsilon^{1}_{ij} + \varepsilon^{1}_{ijk} E_k + \varepsilon^{1}_{ijkl} E_k E_l + \ldots \quad (1)$$

Magnetic phenomena aren't included in classic NLO. It included in parametric crystal optics: Faradey effect and other [2].

Classic NLO phenomenon may be represented as chain of next processes: 1) excitation of corresponding center of light scattering (absorption); 2) change of physical properties of irradiated materials after first process; 3) interaction of light with this excited matter. In the last time we have basic NLO phenomena.

For example, generation of second or third harmonic is connected with two photonic or three photonic absorption of irradiation [1 – 3, 6 – 8].

Phenomena of self-focusing and self-trapping of irradiation are stipulated of nonequilibrium change of irradiated matter (light-induced change of refraction index) [6 – 8]. Difference between times of transmission the basic and self trapping radiation is three order: $10^{-12} – 10^{-11}$ s for basic (linear) radiation and $10^{-9} – 10^{-8}$ s for self-focusing and self-trapping radiation [1, 3].

Difference between generation of harmonics and self-focusing and self-trapping is next. Generations of harmonic are local quantum phenomena, which are stipulated of local excitation of corresponding intrinsic centers of light scattering [1 – 3]. Whereas self-focusing and self-trapping phenomena are the processes of change direction of first order irradiation through the light-induced change of irradiated matter. It is macroscopic phenomenon [1 – 3, 6 – 8].

But both these processes are stipulated of the lifetime of first-excited states. Great value has intensity of irradiation. It must be regime of saturation of excitation of corresponding scattering centers. Other words, next photon must be absorbed of excited center for the time less as lifetime of first order excitation [1 – 3, 6 – 8].

Quantum process (generations of harmonics) are more precisely as macroscopic. Therefore more exacting requirements are presented to the coherence conditions for generations of harmonics. Angle of phase synchronism for generation of second harmonic is few minutes. Whereas self-focusing and self-trapping phenomena are observed in all matters: solid, liquid and gases. Last phenomena are connected of laser-induced breakdown of matter. But it is possible for regime of excitation the matter in the regime of saturation of excitation. Roughly speaking we have inverse excitation of irradiated matter. Self-focusing and self-trapping phenomena are analogous to nonlinear phenomenon of self-blooming phenomenon [2, 5]. But in the last case matter change its properties (conditions of transmission of irradiation) after laser irradiation.

The processes NLO are equivalent to the processes of phase transition (basically second order) [7 – 9]. But for the NLO phenomena these processes are non equilibrium and acted at the time of generation of proper NLO effect.

This analogy NLO phenomena and Landau theory of phase transitions of second order was researched by Y. Haken [8] and developed in [7].

The equivalence between ordered and disordered information may be represented with help de Broglie formula [10], which is used in thermodynamics of point,

$$\frac{S_a}{\hbar} = \frac{S_e}{k_B}, \tag{2}$$

where S_a – action, S_e – entropy, $\hbar$ – Planck's constant, k_B – Boltsman constant.

Formula (2) allows subscribing mutual transitions from quantum to statistical processes. It may be represented as foundation of bond quantum and statistical physics (thermodynamics) [5].

As example theory of second harmonic generation may be represented as phase transition in next way [7, 8].

Noncentral symmetric molecules can interact through a field of transition radiation not only on the frequency of the current field ω , but also on the frequency of radiation each molecule of the second harmonic 2ω [7]. Minimizing the energy of interatomic interaction through field transition radiation at a frequency 2ω in the event of order parameters can also lead to phase transitions. For example, consider the orientation phase transition.

Some substances (liquid crystals, gases, liquids) have the vectors of dipole moment the transition of strongly anisotropic molecules, which are directed along their major axes randomly and distributed in these directions in space. Dipole moments of liquid crystals are oriented in two opposite directions along a particular axis. A similar situation occurs when light isotropic media powerful light waves with linear polarization, when the transition dipole moments are oriented mainly in two opposite directions along the polarization of the wave. However, this orientation does not affect the macroscopic central symmetrical media that appears on its nonlinear characteristics. For example, in systems that are oriented so, the second harmonic generation is impossible.

In the system of molecules, which has powerful coherent light field induced orientation phase transition in which the vectors of dipole moments of transitions of different molecules are oriented in the same direction. It turns out that the system of dipole moments can behave like a ferromagnetic spin systems, with the role of "variables" forces that cause the spin orientation ordering plays an effective intermolecular interaction through field dipole reradiation on frequency 2ω . If the molecules have a permanent dipole moment, whose direction coincides with the direction of the dipole moment of the transition, the orientation on the ordering will coincide well with ferroelectric ordering.

Consider a system of *N* molecules that are locked in a stretched cylinder volume *V* (length along the cylinder axis *z* is equaled *L*), $n = N/V$. We assume that the molecules are highly anisotropic, since the dipole moments of transitions directed along their major axes. In addition, we consider the simple case when the axis of the molecules are oriented primarily along one axis, eg linearly polarized along the axis *x* of the wave:

$$E = e_x \left[E \exp\{-i(\omega t - kz)\} + c.c. \right], \quad |e_x| = 1. \tag{3}$$

Field (3) induces in the molecule, which is at the point r_j , polarization P_j on frequency 2ω , complex amplitude, which can be expressed by a quadratic susceptibility of a single molecule:

$$P_{xj}(2\omega) = \chi_j E^2 \exp(2ikz) \equiv q_j \chi E^2 \exp(2ikz_j),$$

$$\chi = \frac{1}{\hbar^2} \sum_{n,m,l} |d_{mn} d_{ml} d_{nl}| \left[(\rho_{nn} - \rho_{ll}) \zeta(\omega - \omega_{nl}) - (\rho_{ll} - \rho_{mm}) \zeta(\omega - \omega_{ml}) \right] \zeta(2\omega - \omega_{mn}), \tag{4}$$

where $\rho_{\alpha\alpha}$ – population of the respective levels; $\xi(\omega - \omega_{kn}) = \left(\omega - \omega_{kn} + iT_2^{-1}\right)^{-1}$, where T_2 – relaxation time.

In writing (4) uses the fact that the vector of dipole moment passes between levels *m* and *n* of molecule, which is at a point r_j, can be represented in the form $d_{mnj} = |d_{mn}|\hat{e}_x q_j$, where arbitrary number $q_j = \pm 1$, depending on the orientation of the vector d_{mnj}. Further assume that the vectors with different *m* and *n* are collinear.

Average over the ensemble orientational order parameter $\langle q \rangle$ is the number of the examined light-induced transition.

In low light field $\langle q \rangle = 0$. We show that in a sufficiently strong field the phase transition with the appearance of a nonzero order parameter $\langle q \rangle$. To do this, consider that the polarization (4) is a source of field reradiation *j*-molecule on the frequency 2ω. Expressing by solving Maxwell's equations reradiation field at a point r_j through $P_j(2\omega)$, we find the energy of *j*-molecule with other molecules:

$$H_i = -P_i E_i = -4k^2 \sum_{j(j\neq i)} \left[\frac{q_i q_j |\chi|^2 E^4 \exp\{2ik(r_{ij} - (z_i - z_j))\}}{r_{ij}} + c.c. \right], \tag{5}$$

where $r_{ij} = |\vec{r}_i - \vec{r}_j|$.

In the mean field approximation $q_i q_j \to q_i \langle q_j \rangle$, by calculating in (5) the integral over the volume, we are receiving

$$H_i = -2\pi n |\chi|^2 E^4 L k \langle q \rangle q_i. \tag{6}$$

When the light pulse $\tau_i > \tau_r$, where τ_r – the relaxation time of the molecule through orientations, *L* is length (must be more than coherence length). the orientation distribution function has Boltzman form:

$$f(q_i) = C \exp\left(-H_i \Big/ k_B T \right). \tag{7}$$

From condition of self-consistence

$$\langle q \rangle = \sum_{q_i = \pm 1} q_i f(q_i).$$

We receive the formula for order parameter:

$$\alpha = 2\pi n |\chi|^2 E^4 \frac{Lk}{k_B T}. \tag{8}$$

An equation (7) is analogous to equation of magnetization in equilibrium theory of ferromagnetism, but in this case parameter α is depended from intensity of field frequency. Value $\langle q \rangle \neq 0$ is corresponded a case $E^2 > E_{cr}^2$, $\langle q \rangle \to 1$ for $E^2 \to \infty$.

Expanding the right-hand side of (7) in a series of steps *q* we obtain the critical intensity from the condition of transformation to zero a coefficient in the expression for *q*:

$$E_{cr}^2 = \frac{k_B T}{\sqrt{2\pi n |\chi|^2 Lk}}. \tag{9}$$

For $E^2 > E_{cr}^2$ molecules are appeared in system and therefore macroscopic quadratic susceptibility $n\langle \chi_j \rangle = n\langle q \rangle \chi$ occurs, so it becomes possible coherent second harmonic generation.

The physical mechanism of considered light-induced phase transition is as follows. Let in the system of molecules under the influence of coherent light waves, spontaneous (fluctuation) ordering of dipole moments of transitions in the fields is realized to directions $\langle q \rangle \neq 0$. The total field reradiation of these molecules at a frequency 2ω is acting on the nonlinear polarization of the i-molecule at the same frequency 2ω, which is induced by coherent pumping frequency ω, and generates a torque that tends

to return the molecule so as to increase the initial value of the order parameter $\langle q \rangle$. This positive feedback for $E^2 > E_{cr}^2$ leads to instability in the system and the transition to an ordered state for directions d_{mnl}. Note that the occurrence of torque and the corresponding positive feedback and possibly nonstationary case $\tau_i < T_2, \tau_r$. Therefore orientational ordering of the considered case can obviously be under the influence of environment on picoseconds' powerful light pulses.

Thus we considered here light-induced transition due to reradiation field at the second harmonic frequency. However, due to stronger interaction of the field does not lead to orientational phase transition, because $P_j(\omega) \sim q_j^2 = 1,$, that is the interaction energy does not depend on the orientation of the dipole moments of transitions.

Note that registered was the second harmonic generation in a liquid crystal phase. Some scientists suggest the existence of a mechanism for targeting the dipole moments of the molecules in the same direction. Considered this phase transition may be responsible for the observed threshold generation of the second harmonic in the initial media of central symmetrical molecules.

3. PROBLEMS OF RERADIATION AND REABSORPTION IN RELAXED OPTICS

Simple examples of RO processes are the photochemical phenomena. The conditions of effectively of these processes may be divided on two groups [2, 4, 5, 11].

First grope is including the case of light scattering on unstable or metastable centers. For this case basic role have the integral flux of radiation. These processes may be single-photon and cascade multiphoton. The CO_2 – laser annealing of ion-implanted layers $Mg^+/InSb$ in pulse regime (duration of pulse 0,1 mcs) and stationary regime (duration of irradiation 2 s) result to identical results: we have full crystallization of ion-implanted layer and activation of impurity. This process is example of processes of first group.

Second group processes is the processes of light scattering on stable centers. For this case basic role have intensity and time of irradiation. Laser implantation of *InSb* with help Ruby-laser irradiation is example of phenomena of second group.

Basic peculiarities of Relaxed optical phenomena may be analyzed with profiles of laser-induced subsurface donor centers in *InSb* (Fig.1 and Fig.2) [2].

The profiles of the distribution the photostimulated donor centers in subsurface layers *InSb* are represented in Fig. 1 [1, 2]. The samples of p-type conductivity are irradiated by pulses of Ruby laser (wavelength $\lambda = 0,69\,\mu m$, duration of pulse $\tau_i = 20$ *ns*). For intensity of irradiation I_0>0,001 J/cm^2 for *InSb* the n-layers on p-type materials are created. For intensity of irradiation I_0<0,1 J/cm^2 for *InSb* the profiles of the distribution of donor centers are represented the Buger-Lambert law (law of absorption the light in homogeneous media). For further increasing the irradiated intensity the profiles of the concentration donor centers have diffusion nature. The visible destruction of the irradiated semiconductor (melting, the change of the surface colour) had place for I_0>0,3 J/cm^2 for *InSb*.

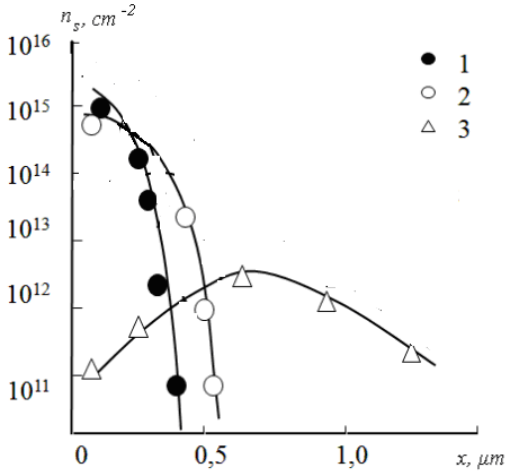

Fig1. *The profiles of the distribution the layer concentration of the donor centers in inverse layers InSb after Ruby laser irradiation with various density of energy (monoimpulse regime): 0,07 (1); 0,1 (2); 0,16 (3)J/cm^{-2}.*

For explanation of results of Fig.1 modified model of photo effect for irreversible case was created [2, 4, 5]. Curves 1, 2 are corresponded to kinetic approximation of this model [2, 4, 5], curve 3 is corresponded to dynamic approximation of this model [2, 4, 5]. Microscopic mechanisms of these results was observed with help model of cascade step-by-step excitation of proper numbers and types of chemical bonds in the regime of saturation the excitation [2, 5, 12]. For indium antimonite two-dimensional lattice of sphalerite was used [12], for silicon – phase diagram [2, 4, 5]. According to this model, curves 1, 2 of Fig. 1 are corresponded to breakage two from three chemical bonds [12]. This case is corresponded of two-photonic absorption and may be represented as irreversible generation of second harmonic, observation of second harmonic for self-absorption range with help of optical and NLO methods is impossible [1].

The dependence of the creation donor centers in subsurface layers of *InSb* after Nd:YAG and Ruby laser irradiation is represented in Fig.2 [13]. The profiles of a distribution of donor centers in $InSb$ after laser irradiation were researched by V. Bogatyryov and G. Kachurin [13]. An irradiation was created with help Ruby laser ($\lambda = 0,69\ \mu m$, $\tau_i = 5 – 6$ ms) and series of pulses Nd:YAG-laser (λ = 1,06 μm, τ_i = 10 *ns*, frequency of repetition of pulses was 12,5 *Hz*). A value of threshold the energy of creation n-layers is equaled ~5 J/cm^2. A tendency of the saturation the layer concentration had place for the energy density ~40 J/cm^2 [13]. These donor centers and proper damages are stable to temperature 400°C [13]. The melting of surface has place for this value of the irradiation [13].

Form of curves 2 and 3 of Fig. 2 showing an influence two- and multiphotonic processes on formation of resulting profile of distribution of donor centers. Subsurface region (~0,5 μm) is corresponded to two-photonic self-absorption as for curves 1, 2 of Fig. 1. Middle parts of curves 2 and 3 of Fig. 2 are corresponded of multiphotonic absorption with photon energy 0,18 eV (band gap of *InSb*). Basic processes for this case are processes of photon fracturing and further reirradiation of bulk semiconductor [5, 11]. Number of reradiations may be 400-500 [5, 11]. Therefore "quantum yield" of creation donor centers for millisecond regime of irradiation is substantially smaller as for nanosecond regime.

Processes of very large laser pumping can cause suppression of oscillation and appearance of chaotization of laser radiation (Haken phenomenon) [2, 5].

All these processes were explained with one physical-chemical point of view, with using elementary energetic estimations [331]. The basic idea of this method is the successive saturation of excitation of proper chemical bonds of irradiated materials [2, 5, 12]. This method allows eliminating differences in the explanation of proper experimental data.

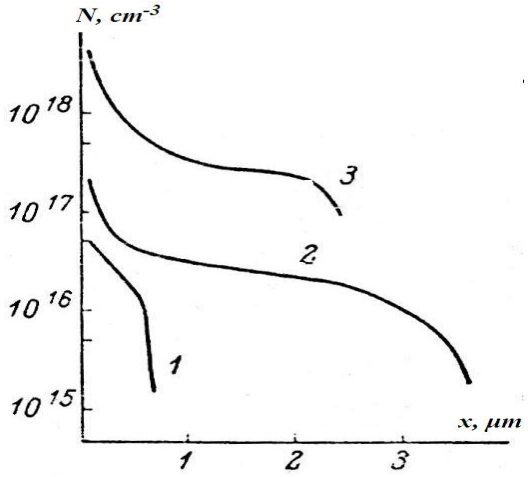

Fig2. *Profiles of the volume distribution electrons after laser irradiation.* 1, 2 – *Ruby laser;* 3 – *YAG:Nd laser. Energy density in pulse, J/cm*^{2}*:* 1 – 5*;* 2 – 40 [2, 5, 13].

For the short regimes of irradiation, when irradiated time is less as relaxation time, the basic processes of irreversible changes in irradiated materials in the regime of saturation of excitations are straight processes of photoinization including multiphotonic processes of absorption. For indium antimonite most probable nonlinear processes for the regime of pulse Ruby-laser irradiation are the photon fragmentation and up-conversion absorption. First effect is basic for the excitation of first chemical bond (one photon break off ~4-5 bonds). This fact is caused grand relaxation time $\sim 10^{-7} s$. Up-

conversion absorption is the result of the scattering Ruby-laser photons on excited electrons of first bond. This effect is caused break off second and third chemical bonds of $InSb$ [2, 5, 12]. For the irradiating time less as first relaxation time $\sim 10^{-7} s$ the processes of irradiated relaxation is negligible. But for the regimes of irradiation with time $\sim 10^{-3} s$ the processes of reirradiation have grand value on the processes of the formation irreversible changes in irradiated materials.

Higher concentration of donor centers for more short regimes of irradiation (Fig.2) is caused of processes of reradiation. For the regimes of irradiation with $\tau_i >> \tau_{r1}$ we have two types of irradiation: first order, basic, Ruby-laser irradiation with $h\nu = 1,78\ eV$, and second order reradiation with $h\nu = E_g = 0,18\ eV$ for $InSb$ [2, 5, 12].

Rough estimation of effects of reemission may be made with help next formulas. The first part of reemission is equaled $I_{r1} = \frac{\tau_{r1}}{\tau_i} I_0 e^{-\alpha x}$. Let this part of absorbed radiation is reemitted. In the next time the absorbed irradiation may be represented in the next form

$$I_{r2} = \frac{\tau_{r1}}{\tau_i}\left(1 - \frac{\tau_{r1}}{\tau_i}\right) I_0 e^{-\alpha x} + \frac{\tau_{r1}}{\tau_i}\left(1 - \frac{\tau_{r1}}{\tau_i}\right) I_0 e^{-\alpha_1 x} I_0 e^{-\alpha_2 x}, \tag{10}$$

where α_1 – absorption factor of radiation with $h\nu = E_g$ (lasing effect) and α_2 – absorption factor of "blooming" radiation.

Second term in (10) is represented up-conversed absorption, which is caused irreversible changes in semiconductor. Second and third relaxation times are considerably greater as time of irradiation. Therefore second term in (10) may be represented as "irreversible" term. For the receiving number of reemission *n* we must multiply second term of formula (10) on *n* and equate to intensity of saturation of excitation I_{sat}. Then

$$n = \frac{\tau_i I_{sat}}{\tau_r\left(1 - \frac{\tau_r}{\tau_i}\right) I_0}. \tag{11}$$

After substitution proper value of I_{sat} from [5, 11] and I_0 from Fig.2 we have $n \cong 10 \div 500.$

It is very rough estimations. But experimental data of Fig.2 are certificated this hypothesis. Surface and volume concentrations donor centers in $InSb$ after irradiation of nanosecond Ruby-laser pulses (Fig.1) is more in 3-4 orders as after millisecond irradiation (Fig.2).

For this case we can propose next simple model. The part of absorbed irradiation with including process of n-reirradiation may be represented as

$$I_n = \frac{\tau_r}{\tau_i}\left(1 - \frac{\tau_r}{\tau_i}\left(1 - \frac{\tau_r}{\tau_i}\left(1 - \frac{\tau_r}{\tau_i}\right)\ldots\right)\right) I_0 e^{-\alpha x}. \tag{12}$$

After using of formula for geometrical progression this relation may be represented in next form [5,11]:

$$I_n = \frac{\frac{\tau_r}{\tau_i}}{1 + \frac{\tau_r}{\tau_i}} I_0 e^{-\alpha x}. \tag{13}$$

With help formula (13) we can receive relation for $\frac{\tau_r}{\tau_i}$ for curves 2 and 3 of Fig.2. In this case we can approximate I_n as average value for tails of this curves and $I_0 e^{-\alpha x}$ as average value for their subsurface parts. For this case we have $\frac{\tau_r}{\tau_i} \approx 0,05 - 0,1.$ For Ruby laser irradiation (curve 2 of Fig. 2.1) we can determine relaxation time $\tau_r \approx (0,05 - 0,1)\tau_i \approx (0,25 - 0,5)\,ms.$ This time is equal zero for curve 1 of Fig.2 because processes of second-order reirradiation have not influence on the irreversible processes.

We can compare the efficiency of generation donor centers for the nanosecond (curve 2 of Fig. 1) and millisecond (curve 2 of Fig. 2) regimes of irradiation. Curve 2 of Fig. 1 is represented pure irreversible process. Therefore the comparative efficiency of using millisecond laser irradiation for creation of irreversible changes in irradiated matter may be determined with help next formula.

$$\beta = \frac{I_{sat} / N_{avsat}}{I_{ir} / N_{avir}}. \tag{14}$$

This value is equaled $2{,}5\cdot10^{-6}$ for subsurface part of curve 2 of Fig. 2.1 and $2{,}5\cdot10^{-7}$ for tail part of curve 2 of Fig. 2.1. For curve 1 of Fig. 2.1 this efficiency is equaled $2\cdot10^{-6}$. Therefore processes of reirradiation may be used for the formation more deep parts of irradiated matter.

Forms of profiles of distribution of donor centers are various too. The maximum of distribution is displaced in volume for nanosecond regime of irradiation. It is effect isn't characterized for millisecond regime of irradiation (Fig.2). Multipulse regime of irradiation of nanosecond Nd: YAG laser is analogous to Ruby-laser in millisecond regime, but for this case we have more discrete process of irradiation. The reemission is caused the decreasing concentrations of donor centers and increasing the depth of donor layers.

Effect of reirradiation in RO is analogous to famous Gamov Urca-process [14] and therefore may be called as optical Urca-process: more part of absorptive and reabsorptive energy is radiated and reradiated and cause not the phase transformations in irradiated matter.

The honeycomb model of laser annealing [15] may be realized in this case too.

These processes were named laser implantation [2, 5]. It may be used for creation new technologies of optical and electronic devices. The problems of a creation the three-dimensional periodical electronic structures are very important and may be having good future.

With help of these processes we can correct proper properties of materials and devices. High thermal stability of receiving donor centers on indium antimonite may be allowed to refine the basic characteristics photo electronic infrared devices. But for this we must select correct regime of irradiation.

This method allows receiving materials with properties, which can't be received with help other methods. Therefore, using of this method is expanded fundamental and applied aspects of modern laser physics and optoelectronics.

4. PROBLEMS OF RERADIATION AND REABSORPTION AS INTERACTION OF NLO AND RO PROCESSES

The general problem of modeling processes of irreversible interactions laser irradiation with solid is very difficult problem [2, 4, 5, 12]. The basic irreversible effects may be having photochemical, thermal or plasmic character [5, 16]. Thermal and plasmic character of this action are determing of collectivization of first-order optical excitations. These processes have various velocities of propagation: thermal – velocity of sound in matter; plasmic – velocity of light in matter. But these processes may be having various actions. Plasmic processes are generated subsurface interferograms and growth of nanohills or nanocolumns. Thermal processes are smoothed and planarized of these structures. As rule, these two processes have mutually opposite directions. Therefore it may be competitive processes. The problem of modeling and observation nonlinear effects in self-absorption range of matter is very difficult problem [5, 8, 17]. Thus we have contradiction: photoionized nature of laser irradiation of semiconductors or other solids and only thermal or plasmic nature of irreversible relaxation of first-order excitations.

Thermal and plasmic processes are the field processes and for the light scattering in matter it are second order processes [5, 16]. Therefore the time of formation of these processes is more as time of first-order quantum processes (photochemical or photocrystall chemical) [2, 4, 5]. Hierarchy of these times is next: time of optical excitation – $10^{-18}-10^{-15}$ s, time of local (quantum) electromagnetic excitation – $10^{-15}-10^{-12}$ s, time of generation of plasmic oscillations – $10^{-13}-10^{-10}$ s, "thermal" times of heating and cooling – $10^{-9}-10^{-5}$ s. First two processes are primary processes, last two processes are secondary.

Primary processes are caused of mechanisms of light scattering, secondary – of relaxation mechanisms. Therefore we must include these results for the explanation of real picture of interaction light and matter.

For bond RO and Nonlinear Optics expansion in series of Pointing tensor (vector) by steps of electric and magnetic fields was used [2, 4, 5]. In this case we have tensor product of electric and magnetic tensors series. This product was used for the classification of proper phenomena. Real part is corresponded to linear and nonlinear optical phenomena, complex part – relaxed optical phenomena[2, 4, 5]. It is allow searching new classes materials with magnetic and electrical properties for observation proper phenomena [2, 4, 5].

RO allowed explaining the role and influence of spectral, time and energy characteristics of laser irradiation on generation of irreversible changes in irradiated matter [2, 4, 5]. This approach was used for the analysis all processes of interaction laser radiation and solid (from luminescence to melting) [2, 4, 5] with help cascade physical-chemical model of excitation in the regime of saturation.

Interference and diffractive phenomena of RO may be observed with help plasmic models [2, 4, 5]. Circular and elliptic polarizations of irradiation allow generating homogeneous surface nanostructures. Here height is changed from 15-100 nm for nanosecond regime of irradiation Fig.3 [18] to 400-450 nm for femtosecond regime Fig.4 [19]. Parameters of irradiation for Fig. 3 are next: pulse duration 15 ns, wavelength 1,06 μm, pulse rate 12,5 Hz, power 1 MW [18].

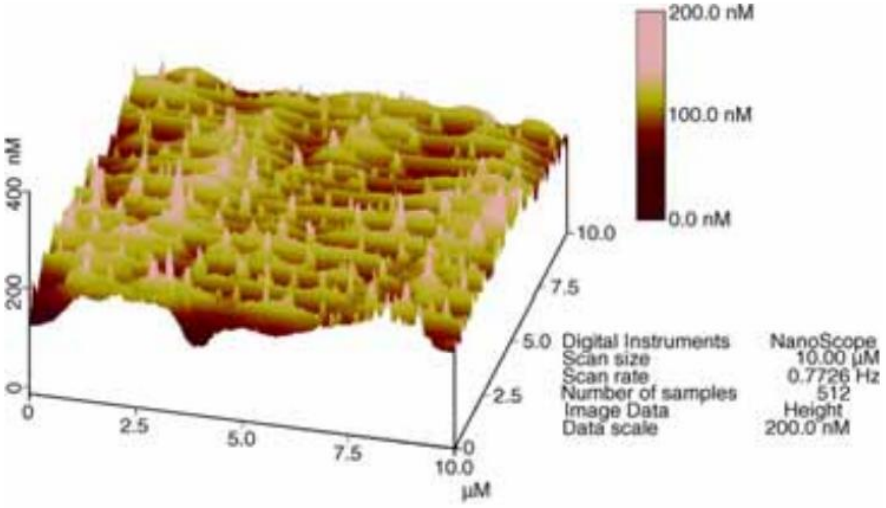

Fig3. *Three-dimensional AFM image of self-organized nanostructures formed under Nd:YAG laser radiation at intensity of 28 MW/cm^2 of Ge surface* [18]

Height of surface nanostructures for the nanosecond regime of irradiation is maximal (100 nm) for the germanium [18]. For the silicon, gallium arsenide and metal films high of laser-generated surface nanostructures is change from 10 nm to 20 nm. This difference can be explained in next way. Index of absorption of Ge crystal of diamond symmetry is more as silicon with this symmetry. But surface part of irradiated germanium is transited to hexagonal phase. It is experimental data. Other result given's phase transitions. The hexagonal lattice of germanium has greater size as diamond modification. Therefore hexagonal nanostructures are greater and more stable as polycrystalline or metallic nanohills.

Mechanisms of creation other laser-induced nanostructures may be explained on the basis cascade model of step-by-step excitation of corresponding type and number of chemical bonds in the regime of saturation of excitation. According to this model decrease of symmetry of irradiated matter is occurred with increase of intensity of irradiation (case of Ruby and Nd laser irradiation of silicon, germanium and carbon) [2, 5].

But in [18] explanation of creation laser-induced hexagonal phase on germanium surface is based on the Bernar phenomenon: generation of hexagonal phases in heated liquid on roaster. This effect is observed for few liquids. Chandrasekar theory is described this process as thermal-diffusive processes [8, 20, 21]. In this case we have transition from more low volume symmetry to more high surface symmetry. Chandrasekar created the hydromagnetic theory of creation sunspots [20].

For laser-induced creation volume hexagonal structures on *Ge* we have inverse transition: from high volume symmetry (diamond modification) to more volume low symmetry (hexagonal symmetry) [18].

Now we represented estimations of process the creation laser-induced hexagonal structure on the basis Chandrasekar-Haken theory [8, 20, 21] and cascade model [2,5]. Creation of instability is characterized of critical point. This point my be characterized by Rayleygh anf Nusselt numbers [20, 21].

Rayleygh number is determined as

$$Ra = \frac{g\alpha\beta}{\kappa\nu}h^4, \tag{15}$$

where g – free fall acceleration, α coefficient of volumetric expansion, $\beta = \frac{\Delta T}{h}$ – temperature gradient, κ – coefficient of heat conductivity, ν – kinematic viscosity, h – thin of heated films.

Nusselt number determined as

$$Nu = \frac{|q|h}{\kappa_0 \Delta T} = \frac{|q|}{\beta\kappa_0}, \tag{16}$$

where κ_0 – statistical value of coefficient of heat conductivity, q – full heat flow.

$Nu = 1$ for case of heat transfer only with help heat conductivity and $Nu > 1$ for case of heat transfer with help heat conductivity and convection. Behavior function $Ra(Nu)$ in critical point is analogous to curve of phase transition.

Critical value Rayleygh number is equaled $Ra_{crit} = 1700 \pm 51.$ For $Ra > Ra_{crit}$ $Nu > 1$. For regime of irradiation of Fig. 3 $Ra < 1$ and $Nu < 1.$ Therefore application this theory to these results is very ambiquous and discussed.

According by Haken probability of creation hexagonal structures is major for $Ra \geq Ra_{crit}.$ For further increasing of Rayleygh number a generation of cylindrical structures is basic. It explains of occurrence the curls in atmosphere [8].

But conditions of creation new phases in solid phase is other as in liquid phase. Chandrasekar – Haken theory may be used for the modeling processes of growth $Si_{1-x}Ge_x$ whispers with diameter > 40 μm [22]. These crystals have hexagonal cross-section. Basic methods of receiving these structures are thermal (epitaxial and sputtering, including laser ablation)). For decreasing sizes to 1 μm we have circular cross-section [22] and properties of whispers is identical to bulk matter. For case of laser implantation (Fig. 3 – Fig. 6) we must include chain electromagnetic processes of creation vortexes (nanohills and nanocolumns) and chain photochemical processes, which are connected with intensive photo ionization of irradiated layers.

More pure ionizing results were received for the irradiation silicon by femtosecond laser pulses (Fig. 4 – Fig. 6) [19].

More real is explanation of results of Fig. 3 and Fig. 4 – Fig. 6 on the basis cascade model of excitation of corresponding chemical bonds in the regime of saturation of excitations. Density of energy of irradiation was 0,42 J/cm^2 (experimental data). Density of energy, which is necessity for the one bond breakage for Fig. 3, is equaled ~ 0,1 – 0,2 J/cm^2. If we allowed the reflection factor and fact that irradiation was realized in focused regime then last value must be ~ 0,2 – 0,4 J/cm^2. This value is coincided with experimental data. Therefore basic mechanism of creation hexagonal structure of Ge on diamond structure of this material is photochemical.

Irradiation of SiO_2/Si layer by second harmonic of Nd:YAG laser (wavelength 532 nm) with density of power 2 MW/cm^2 or density of energy 0,03 J/cm^2 is generated nano-hills with height 10-15 nm without change of crystal symmetry [18]. For the creation hexagonal structures of silicon the density of energy must be 0,47 – 0,71 J/cm^2. With including reflection factor and focusing character of irradiation last values must be 0,9 – 1,4 J/cm^2. Therefore creation of hexagonal structure of silicon for this regime of irradiation is impossible.

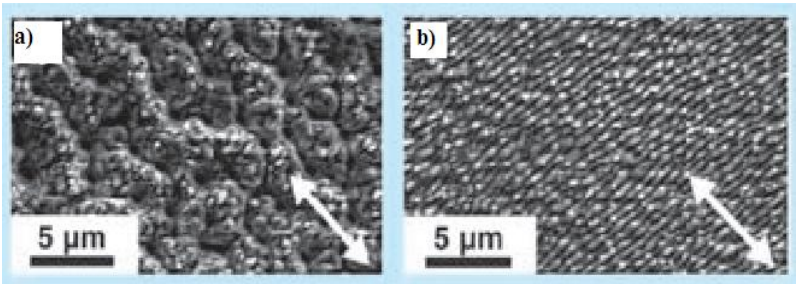

Fig4. *Ordered structures, which were generated on surface of silicon after laser irradiation through lay of water, (arrow in lower angle show the direction of polarization of laser radiation); duration of pulse 100 fs, number of pulses – 200, wavelength – 800 nm, density of energy the irradiation a) 0,25 J/cm^2, b) 0,05 J/cm^2* [19].

Other picture we have for the femtosecond regime of irradiation (Fig. 4 and Fig. 6) [19]. It is results of second-order irradiation of first order irradiated silicon (duration of pulse 100 ns, wavelength 800 nm, number of pulses 200, density of energy of irradiation 2,5 and 0,5 J/cm^2, Fig. 4)). Estimations of cascade theory gives values ~1,5 – 2,0 J/cm^2. With reflectance these values are equaled ~2,0 – 2,7 J/cm^2 [2, 5]. In this case we have possibility of creation hexagonal structures. It certifies of large height of nanocolumns (400 – 450 nm, Fig. 5).

But in this case we have influence of collective electromagnetic fields on formation finished structures too. Therefore the threshold of creation new phases with low symmetry as initial may be less as for pure photochemical regime of irradiation and probability of creation of other more low crystal and quasi crystal phases is increased for motion by one nanocolumn from its basis to peak (end). It may be explained the relative large value of nanocolumns height.

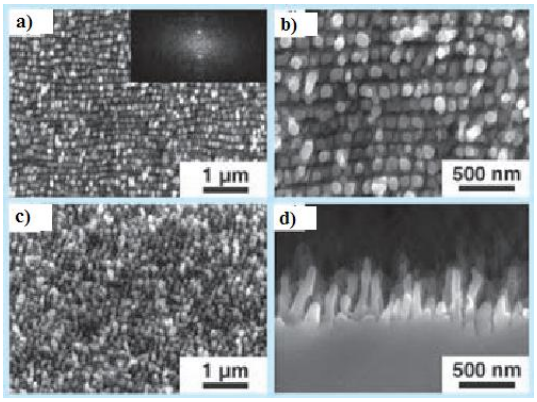

Fig5. *Nanocolumns, which are generated after irradiation structures of Fig. 4, (wavelength of irradiation 800 nm, number of pulses – 200, density of energy of irradiation 0,5 kJ/m^2): a) and b) turn of polarization on* 90°*, b) turn of polarization on* 45°*, d) cross chip of nanocolumns. On insertion to Fig. 5a – Fourier-picture of structures* [19].

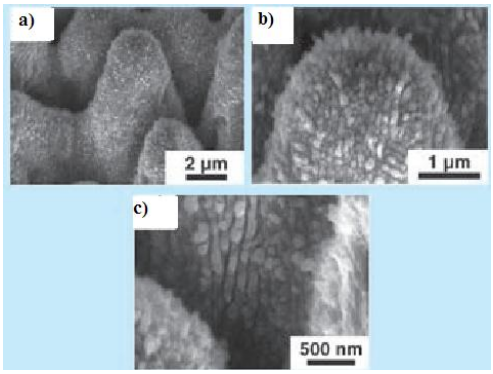

Fig6. *Surface nanocolumns of little scale, which have orthogonal orientation to a crests of nanorelief of large scale* [19].

Surface field distribution of energy of irradiation gives Makin plasma model [19], which is based on interference of fall radiation with laser-generated surface polariton-plasmons and on interference between surface polariton-plasmons. These polariton-plasmons have non equilibrium and may be have irreversible nature. Resulting nonostructures are the results of these interactions. It may be

interference between fall radiation and laser-induced frozen picture, that was created in the time of previous irradiation. Roughly speaking, sequential irradiation is caused a generation step-by-step set of structures. These results are illustrated on Fig. 6. Polariton-plasmon model shows the influence of polarization the irradiation on the processes of formation final interference patterns [19].

Creation of non equilibrium and irreversible polariton-plasmons may be explained of basis of laser-induced swelling of surface. Surplus of laser-generated negative electrical charge in surface and subsurface regions of irradiated matter is generated swelling of surface. This swelling has electromagnetic nature. Thermal processes have diametric inverse direction. Verification of this concept is experimental data of Fig.6 [19]. Second order laser-induced nanostructures are created in direction, which is perpendicular to irradiated surface.

These models may be used for the explanation of the fabricate SiGe nanowires using pulsed ultraviolet laser induced epitaxy [23]. In this case heterostructure Si (depth 12 nm) – Ge (depth 6 nm) were irradiated spatially homogenized 308 nm XeCl ecimer laser beam with pulse duration 27 ns with density of energy 0,69 J/cm^2. This regime of irradiation may be caused the melting of irradiated layers and creation new *Si – Ge* structures. The duration of melting pulse is 26 ns and this pulse is retarded relatively to irradiated pulse on 15 – 20 ns. This pulse must be consisted from two pulses for *Ge* and *Si*. Only subsurface region of *Ge* is mix with surface layer *Si*. Basic mechanisms of creation SiGe structures is diffusive. Absorptance of Ge is 10^5 cm^{-1}, silicon - $3\cdot10^4$ cm^{-1}. Therefore melting layer is heating more quickly for *Ge* as for *Si* and this fact is caused the motion germanium layers to up and silicon layers to down. As result we have *SiGe* structures [13]. Small region of absorption and localization the laser radiation is caused the creation this nanowires.

In [24, 25] for minituarization of receiving structures of crystals 4H-SiC were irradiated by pulses of femtosecond laser (duration of pulses 130 *fs*, wavelength 800 *nm*, frequency of pulses 1 *kHz*, density of energy 200-300 *nJ*/pulse) with help microscope.

Conditions of irradiation are represented in Fig. 7 ((a), (b)) [24]. Femtosecond laser pulses were irradiated along the lines inside 4H-SiC single crystals at a depth of 30 μm by moving the sample at a scan speed of 10 $\mu m/s$. The laser beam was irradiated at a right angle to the to the (0001) surface of the crystal. The irradiated lines were almost parallel to the $[1\,\bar{1}00]$ direction. A schematic illustration of the laser-irradiated pattern is shown in Fig. 7 (a). The distance between neighboring lines was 20 μm.

Bright-field TEM (transmission electron microscopy) image of the cross section of a line written with a pulse energy of 300 *nJ*/pulse is shown on Fig. 7 ((c) – (e)) [24].

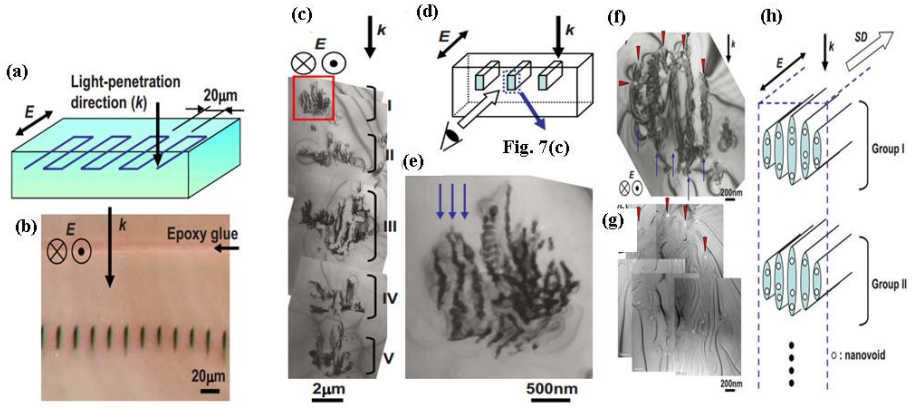

Fig7. *(a) Schematic illustration of the laser irradiated pattern. The light propagation direction (k) and electric field (E) are shown. (b) Optical micrograph of the mechanically thinned sample to show cross sections of laser-irradiated lines (200 nJ/pulse). (c) Bright-field TEM image of the cross section of a line written with pulse energy of 300 nJ/pulse. (d) Schematic illustration of a geometric relationship between the irradiated line and the cross-sectional micrograph. (e) Magnified image of a rectangular area in (a). Laser-modified layers with a spacing of 150 nm are indicated by arrows. (f) Bright-field TEM image of a portion of the cross section of a line written with a pulse energy of 200 nJ/pulse. (g) Zero-loss image of a same area as in (f) with nanovoids appearing as bright areas. Correspondence with (f) is found by noting the arrowheads in both micrographs. (h) Schematic illustrations of the microstructure of a laser modified line. Light-propagation direction (k), electric field (E), and scan direction (SD) are shown. Only two groups (groups I and II) of the laser-modified microstructure are drawn.*

In contrast to the formation of surface periodical structures three-dimensional periodic structures were obtained in this case. Sectional area of these structures was ~ 22 μm, the depth of ~ 50 μm. As seen from Fig.7(c) we have five stages disordered regions, which are located at a distance from 2 to 4 μm apart vertically [24]. Branches themselves in this case have a thickness from 150 to 300 *nm*. In this case there are lines in the irradiated nanocavity spherical diameter of from 10 *nm* to 20 *nm*. In this case irradiated structures have crystallographic symmetry of the initial structure.

More detail information about processes, which are generated in first two stages, represents in Fig. 7 ((f) – (h)) [25].

Explanation of the experimental data, which are shown in Fig. 7, is based on nanoplasmic model [24, 25] . The emergence nanovoids explained on the basis of the explosive mechanism. However, the same result can be explained by the formation of vacancy clusters, especially those sizes of nanovoids same are equivalence to sizes of nanoclusters. Nanovoids, as a rule, are formed between the most modified regions, i.e. in these areas there are sinks of vacancies [26], which form the nanovoids or vacancies clasters.

This is due to redistribution of charge (ions) for the ionization of solid state, which leads to a redistribution of major semiconductor components [2, 5]. Volume periodicity of structures may be explained on the basis of change of absorption conditions in irradiated matter, therefore the radiation begins to focus in the deeper areas. It may be explained with help Lugovoy – Prokhorov or generalizing Lugovoy – Prokhorov theory of moving focuses. Thus we have irreversible trace of moving focuses in matter. In classical NLO this trace has nonequilibrium nature. A creation of "fiber" patterns (Fig. 7(c), Fig. 7(e)) is caused of ionizing optical breakdown of irradiated matter. It is trace of filament of matter. In 4H-SiC this filament has size few micrometers. In air it has size few hundred meters. This phenomenon is analogous to creation of streamers [27] or formation of lightning [5].

Physical-chemical transformations in more deep layers are generated with help multuphotonic processes of absorption with more large number of laser pulses as in surface and subsurface layers [5].

But this model gives not physical and chemical properties of created nanostructures. These questions may be resolved with help cascade model of excitations of corresponding chemical bonds in the regime of saturation of excitation.

5. CONCLUSIONS

- Problems of reemission and reabsorption in Nonlinear Optics are analyzed on examples generation second harmonic and phenomenon of self-focusing and self-trapping is discussed.
- Analogy between phase transitions and nonlinear optical processes is observed too on the example of generation second harmonic.
- The conditions for laser annealing of ion-implanted layers and laser implantation are formulated on the basis of laser annealing Mg^+/InSb layers and InSb.
- Criteria of reradiation and its influence, with help multiphotonic absorption, on the generation RO processes and phenomena are formulated and used.
- Chandrasekar-Haken theory of Bernar phenomenon is analyzed and we show that this theory may be used for the modeling processes of creation structures with sizes > 40 μm.
- Concept of polariton-plasmon model and laser-induced swelling of irradiated surface must be used for the explanation of creation surface interferograms and nanostructures.
- Laser-induced phase transformations were observed with help cascade model of step-by-step excitation of corresponding type of chemical bonds in the regime of saturation of excitation.
- Creation of periodical laser-induced volumetric micro and nanostructures was explained on the basis of nanoplasmic model, Lugovoy – Prokhorov theory of moving focuses and cascade model of excitation of proper chemical bonds in the regime of saturation of excitation.

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