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Wave-like destruction on the input surface of optical media by powerful nanosecond laser pulses as a manifestation of stimulated radial scattering in the surface layer

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The wave-like damage structures on the entrance surface of optical media, which observed in various experimental studies with nanosecond laser pulses, are analysed. Sometimes such structures appear as a system of straight lines intersecting at the an-gles defined by the crystallography of a sample. It is proposed that stimulated radial scattering in a surface layer of a sample is responsible for their formation.

Keywords: stimulated radical scattering, wave beating in the surface layer, plastic deformation.

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1. Introduction

Wave-like damage formed on the surface of various media in the form of periodic waves or a system of concentric rings as a result of powerful laser exposure is observed in various experimental studies. As a rule, the greatest interest, both for theorists and experimenters, are waves on the surface of metal films formed under the action of femtosecond pulses [1–3]. Their formation is associated with plasmon-polariton scattering from the boundaries of surface inhomogeneities, as a result of which the surface wave periodically interacts with the pump wave at the points of coincidence of their phases, forming standing waves, in the beams of which energy is released, leading to distortion of the surface. The release of energy leading to ablation of the medium during plasmon-polariton scattering occurs due to the scattering of interacting waves on free electrons formed due to multiphoton ionization. The high flows required for this ($I \sim 10^{13}\,\mathrm{W/cm^2}$) are implemented with femtosecond pulses. Since in this case there is no difference in the frequency distribution of the interacting waves, the surface waves are formed with a period close to the period of the light wave.

In the present study, waves are considered not on the metal surface, but on the input surface of transparent optical media formed due to phonon-polariton scattering in the surface layer, which in fact is Raman scattering, the release of energy at which occurs due to the difference energy between the Stokes wave and the pump wave, which goes to the resonant excitation of molecular vibrations, and occurs at much lower intensity nanosecond pulses ($I \sim 10^8-10^9\,\mathrm{W/cm^2}$). As a source of Stokes waves propagating along the surface, a mechanism for their formation in transparent dielectrics is proposed, based on the manifestation of radial scattering [4] in the surface layer. As a result, the Stokes waves radially scattered from the center or inhomogeneity boundary running along the surface interact orthogonally with the pump wave falling to the surface at the points of their beats. This makes the period of the resulting waves much longer than the period of the pump wave, and the strengthening of Stokes waves in the process of resonance interaction and excitation of new phonons significantly slows down the process of their attenuation. As a result, we form weakly attenuated waves with a large number of periods. In principle, this is the result of field interference created by phonon-polariton scattering in a surface layer with an orthogonally incident pump wave field.

In the previous study [4] surface craters with a wavy surface were described, formed not as a result of ablation, but as a result of plastic deformation of the medium under the influence of interference of a periodic sequence of shock waves formed during the joint development of Brillouin and Raman scattering. No additional scattering that could be associated with avalanche ionization (optical breakdown) was detected in the study [4]. In the study [4] it has been shown that such a mechanism, when scattered on micro-heterogeneities, leads to the formation of longitudinal-radial periodic waves, the radial period of which is determined by the period of the Stokes scattering wave from the center to the outside. As a result of their orthogonal interaction with the pump wave, we form shock waves converging on the axis.

The seeds for the development of both longitudinal and radial scattering are any inhomogeneities in the optical medium, in which the binding energy of the atoms that form them differs from the binding energy of atoms in a homogeneous medium. This leads to the excitation of oscillations of atoms at their resonant frequencies in the field of the pump wave, and, consequently, a change in the

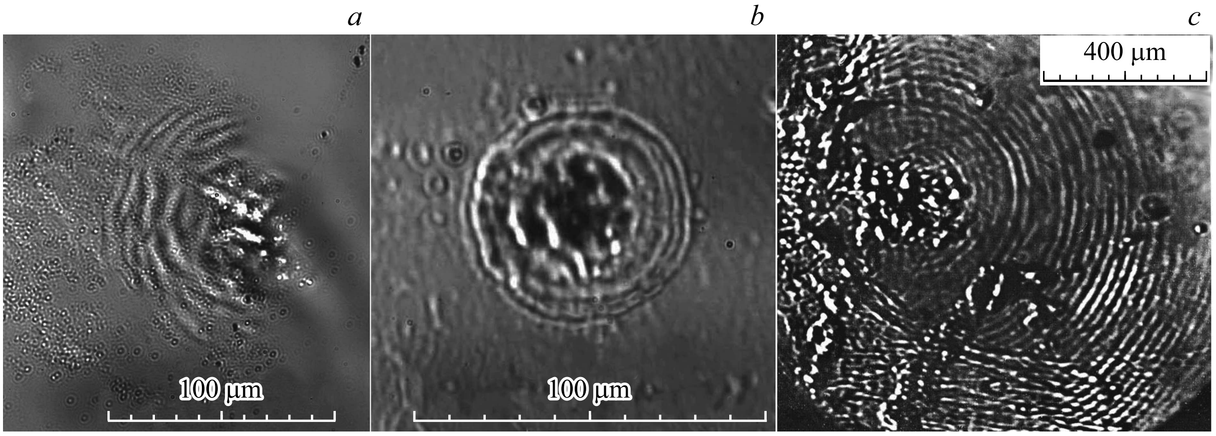

Figure 1. A ring structures: $a{-}b$ — on YAG surface when exposed to a laser on the color centers ($\tau \sim 20$ ns, $\lambda \sim 920$ nm, $I \sim 5 \cdot 10^9$ W/cm^2), and c — on surface Ge ($\tau \sim 1\,\mu$s, $\lambda = 10.6\,\mu$m, $I \sim 4 \cdot 10^8$ W/cm^2).

frequency of the photons scattered on them, which means the release of energy. Stokes scattering waves at sufficient pumping intensity lead to the excitation of both Brillouin scattering towards the pump wave with the formation of acoustic waves, and Raman scattering. In the event that we simultaneously develop the Mandelstam–Brillouin scattering, which, with a long pulse duration, leads to the germination of acoustic waves towards the pump wave to the surface with a simultaneous increase in the scattering intensity, and in the maxima of acoustic waves Raman scattering occurs, then due to the large pulse transmitted to the atoms of the medium, their transformation into a periodic sequence of shock waves occurs. At high intensity of these waves, they can lead to the breakthrough of the surface described in the study [5], which is most pronounced in media with a low phase transition threshold. Though the Stokes Raman scattering can simultaneously occur in the transverse (radial) plane, in which, orthogonally interacting with the traveling pump wave at the points of coincidence of their phases, excite stimulated radial scattering. In this case, the addition of their pulses leads to the formation of a periodic sequence of shock waves converging on the axis at an angle of $\sim 45^\circ$, causing a sharp increase in temperature and pressure on it. As a rule, there is a simultaneous development of longitudinal and radial scattering. Since shock waves converge on the axis in the form of concentric waves, in optical media with a low phase transition threshold (YAG, Si, Ge, AsGa) they can form symmetrical push holes (microcraters) of small size on the surface $\varnothing \sim 3{-}10\,\mu$m, shown in the study [4].

In this study, we will consider the mechanism of formation on the surface around the formed crater of concentric waves with a constant or continuously changing radius period [6]. Similar concentric waves on the surface of the slopes of the resulting crater are observed around its center [7], which is, as a rule, the center of radial scattering in the form of Stokes waves. Sometimes concentric waves are bordered by a system of parallel lines intersecting at certain angles.

In this study, an attempt is made to explain the mechanism of their formation by radial scattering.

2. Excitation of concentric surface waves

In the study [4] the mechanism of formation of radial waves in the case of a flat wavefront of the pump wave scattered on microheterogeneities of the medium was described. If this process led to the germination of radial waves from the seed heterogeneity to the input surface, then a system of concentric rings with different periods sometimes arose around the crater formed on the surface with sharp boundaries (at an increased intensity at which the threshold is exceeded) (Fig. 1, $a{-}c$). Fig. 1, $a{-}b$ shows the ring structures on the surface of the YAG crystal, when pumped with a nanosecond pulse with a wide spectrum, obtained at the installation described in the study [4].

The structures on the Ge surface, shown in Fig. 1, c, are obtained by pumping a microsecond CO_2 laser from the study [6]. These figures, against the background of the rings, show some individual microwells of pushing. Surface waves develop from boundaries, which are the source of scattering of Stokes waves propagating along the surface, on which the pump wave interacting with them perpendicularly falls. These waves, evolving from boundaries along the surface, track the curvature of these boundaries.

This is clearly shown in Fig. 2 from the study [8]. In Fig. 2, a you can see periodic waves running parallel to the cracks formed on the surface of the crystal S–FAP — [$Yb^{3+}:Sr_5(PO_4)_3F$] after ablation of the layers of the anti-reflective coating ($HfO_2{-}SiO_2$). It can be seen that in the case of blurring of the boundaries of waves comparable to the period (vertical crack on the right), waves do not form. A fairly clear system of periodic waves, the bends of which reflect the bends of the crater boundaries, is presented in

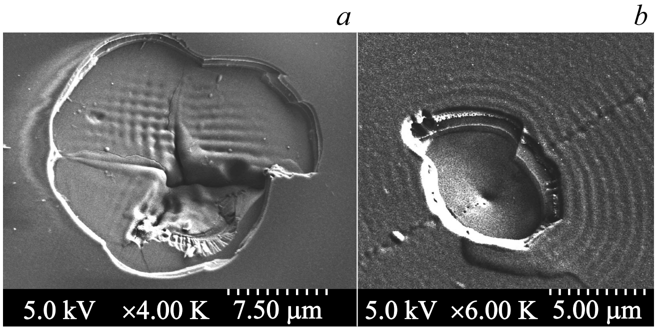

Figure 2. a — waves on the surface of S−FAP after ablation of the anti-reflective layers from it, b — periodic waves on the surface of the anti-reflective layers developing from the boundaries of the crater formed as a result of ablation of these layers ($\tau \sim 3.5$ ns, $\lambda \sim 1064$ nm, $I \sim 5.7 \cdot 10^9$ W/cm^2).

Fig. 2, b, which was observed on the surface of the anti-reflective coating applied to the surface S−FAP. The sample was exposed to radiation from Nd:YAG laser at wavelength of 1064 nm with a duration of 3.5 ns and an energy density of 20 J/cm^2.

Radial waves developing inside the optical medium themselves choose the angle at which they will be in synchrony with the pump wave. In the medium, the interaction of the Stokes scattering wave and the pumping traveling wave occurs at the points of coincidence of their phases. Since the period of the Stokes wave (T_{st}) is longer than the period of the pump wave (T_p), the coincidence of their phases occurs with a shift from the vertical (from perpendicular to axis). This shift in the longitudinal direction over the period of their interaction, expressed through their periods, is equal to $T_{\text{st}} - T_p$. This difference divided by the period of the Stokes wave (a right triangle with a hypotenuse equal to T_{st}) determines the angle of inclination of the Stokes wave with respect to the perpendicular to axis

$$\sin\alpha = \frac{T_{\text{st}} - T_p}{T_{\text{st}}}.$$

Since the periods of the Stokes wave and the pump wave are equal

$$T_{\text{st}} = \frac{\lambda_{\text{st}}}{cn_{\text{st}}} \text{ and } T_p = \frac{\lambda_p}{cn_p}, \text{ so} \alpha = arc\sin\left(1 - \frac{\lambda_p n_{\text{st}}}{\lambda_{\text{st}} n_p}\right).$$

This angle automatically adjusts to the change in the intensity of the interacting waves that change the refractive index of the medium.

Stokes waves scattering from the boundaries of inhomogeneities along the surface do not have such a possibility, and the coincidence of their phases with the phase of the pump wave falling normally on the surface occurs at the points of beat of these waves. The frequency of wave beats ν_{be} is equal to the difference between the spatial frequencies of $\nu_{be} = \nu_{\text{st}} - \nu_p$, the Stokes wave in the medium ($\nu_{\text{st}} = n_{\text{st}}/\lambda_{\text{st}}$) and the pump wave ($\nu_p = n_p/\lambda_p$). At the same time, each maximum wave of beats is modulated with a frequency

$$\nu = \frac{\nu_{\text{st}} + \nu_p}{2}.$$

The value inverse of the frequency of the beats

$$\frac{1}{\nu_{be}} = \lambda_{be} = \frac{1}{\frac{n_p}{\lambda_p} - \frac{n_{\text{st}}}{\lambda_{\text{st}}}}$$

determines the period of the surface waves formed.

As a result of the orthogonal interaction of these waves, which occurs when the difference in their phases is $2\pi m$, where m is integer, we get a standing surface wave, the period of which is equal to the period of beats of the interacting waves. The initial phase of the Stokes wave is equal to the phase of the pump wave at the time of its scattering, therefore, with a coherent pump wave, their phases retain a mutual correlation. In principle, this is also Raman scattering in the transverse or radial direction, only not within the medium, but in its surface layer, in which the wave running along the surface and interacting with the pump wave is a Stokes wave of Raman scattering. This can be called a stimulated phonon-polariton radial scattering at the interface of two media, obeying dispersion relations different from normal dispersion.

That is, the pump waves scattered on the phonons excited by radial scattering already in the form of Stokes waves themselves interact with the pump wave in the surface layer at the points of coincidence of their phases. But in order for surface waves to form, it is necessary that the source of Stokes surface scattering has a sharp fixed boundary. The resulting transfer of difference energy and momentum to surface molecules leads to the formation of surface waves. Since the initial phase of these waves does not change (the boundaries of the hot regions are considered stationary), the points at which the waves interact to form phonons and Stokes photons will be stationary. In the case of displacement of the boundaries (boundaries of the crater) in the process of exposure to the pumping impulse, the maxima of the generated waves are also shifted by the same amount.

To determine the period of the formed surface waves, it is necessary to know the refractive index for the Stokes wave and the nature of its change. The dielectric constant is generally equal to

$$\varepsilon = \varepsilon_0(1 + N\alpha) = \varepsilon_0\left\{1 + N\left[\alpha_0 + \left(\frac{\partial\alpha}{\partial X}\right)_0 X\right]\right\},$$

where N is concentration of the oscillators involved in light propagation, and $(\partial\alpha/\partial X)_0$ is differential polarizability. The maximum change in dielectric polarizability due to the resonant interaction of the waves is related to the nonlinear term

$$\varepsilon_0 N\left(\frac{\partial\alpha}{\partial X}\right)_0 X.$$

To determine it, it is necessary to obtain an expression for the normal oscillatory coordinate of oscillators, which can be obtained from the macroscopic theory of forced Raman scattering, developed in Yariv's classical study [9]. The above study uses a model of a scattering medium consisting of N harmonic oscillators (per unit volume), each of which corresponds to one molecule. Oscillators are independent of each other, i.e., an ensemble of oscillators cannot support wave motion with a group velocity other than zero. Each oscillator is characterized by its position of z and a normal oscillatory coordinate $X(z,t)$. From solving the equation of motion for one oscillator, from the study [9] for the case of radial scattering we get

$$X(r,t) = \frac{\varepsilon_0 \left(\partial\alpha/\partial X\right)_0 E_p E_{\text{st}}^* e^{i[\omega_0 t - (k_p - k_{\text{st}})r]}}{8m[\omega_0^2 - (\omega_p - \omega_{\text{st}})^2 + i(\omega_p - \omega_{\text{st}})\gamma]} + \text{k.s.},$$

where E_p and E_{st} — the field of pump wave and Stokes wave, ω_p and ω_{st} — their frequency, and γ — constant attenuation. For our case, $\omega_0 = \omega_p - \omega_{\text{st}}$, where ω_0 is the resonant frequency of the oscillator, the expression for the coordinate is converted to

$$X(r,t) = -i\,\frac{\varepsilon_0 \left(\partial\alpha/\partial X\right)_0 E_p E_{\text{st}}^* e^{i[\omega_0 t - (k_p - k_{\text{st}})r]}}{8m\omega_0\gamma} + \text{k.s.}$$

The oscillation phase of molecules $(k_p - k_{\text{st}})r$ is not a consequence of wave propagation at a frequency of ω_0, but reflects the fact that the vibrations of molecules are excited by the product of electric fields with frequencies ω_{st} and ω_p and, therefore, the phase of these oscillations is determined by the excitation force phase $E_p E_{\text{st}}^* e^{i(k_p - k_s t)r}$. Since the spatial position of the maxima is determined by the difference in wave numbers: $(k_p - k_{\text{st}})r$, then $(k_p - k_{\text{st}})^{-1}$ defines their period, from where we determine the distance between them equal to the inverse frequency of the beats of the interacting waves:

$$\lambda_{be} = \frac{2\pi}{k_p - k_{\text{st}}}.$$

Since the oscillators (phonons) are independent, their energies are quantized, and the group velocity is zero, the expression for the oscillator amplitude $X(r,t)$ shows the distribution of their density $N(r)$. Substituting an expression for $X(r,t)$ into an expression for the dielectric constant, we obtain

$$\varepsilon = \varepsilon_0(1 + N\alpha) = \varepsilon_0(1 + N\alpha_0) + \left[-i\varepsilon_0^2 N \left(\frac{\partial\alpha}{\partial X}\right)_0^2 \frac{E_p E_{\text{st}}^* e^{i[\omega_0 t - (k_p - k_{\text{st}})r]}}{8m\omega_0\gamma} + \text{k.s.}\right],$$

which can be represented as $n^2 = n_1^2 + n_2^2$, where $n_1^2 = \varepsilon_0(1 + N\alpha_0)$, and

$$n_2^2 = -i\varepsilon_0^2 N \left(\frac{\partial\alpha}{\partial X}\right)_0^2 \frac{E_p E_{\text{st}}^* e^{i[\omega_0 t - (k_p - k_{\text{st}})r]}}{8m\omega_0\gamma} + \text{k.s.}$$

Thus, the n_2^2 is proportional to the product of the interacting fields: $n_2^2 \propto E_p E_{\text{st}}^* e^{i[\omega_0 t - (k_p - k_{\text{st}})r]}$, and changes continuously, tracking the beats of the waves. That is, this product of the fields determines the distribution of the concentration of resonantly excited molecules in the surface layer.

Thus, any change in the intensity of the pump wave caused by its radial heterogeneity, or the intensity of the Stokes wave, which can also be associated with its radial weakening when moving away from the source, leads to a change in the refractive index. Despite the fact that we have a resonant excitation of molecular oscillations (oscillators), this does not lead to a singularity with the reflection of the pump wave from the surface, but there is an intensive pumping of energy from the pump wave into the Stokes wave propagating along the surface, occurring with a significant increase in the refractive index in the maxima of the wave beats. Thus, the refractive index propagating along the surface of the Stokes wave will periodically change from its minimum value in the minimums of surface waves to the maximum value in their maxima, in which its maximum interaction with the pump wave occurs. Due to the orthogonal interaction of the pump wave and the Stokes wave, the excitation of molecules occurs with the transfer to them of the total momentum $p = \hbar(k_p + k_{\text{st}})$, which is directed at an angle $\sim 45^\circ$ to the surface, causing its plastic deformation in the interaction maxima and ablation.

To determine the period of surface waves, it is necessary to know the refractive index for the Stokes wave, while the refractive index for the pump wave acting on the surface with a phase coinciding with the Stokes wave and a constant amplitude does not affect the period of excited oscillations in the surface layer, and it can be taken to be equal to one. We can make our lives easier by believing that we know the refractive index averaged by the period of the waves formed and its dependence on intensity. In this case, it is possible to qualitatively show the dependence of the period of maxima of interacting waves on the Stokes shift and the angle at which they interact along the surface.

As shown above, the distance between the maxima of the resulting wave along the surface is

$$\lambda_{be} = \frac{1}{\dfrac{n_p}{\lambda_p} - \dfrac{n_{\text{st}}}{\lambda_{\text{st}}}}.$$

This formula, due to the uncertainty of the values of the refractive index, does not allow to accurately determine the period of the surface waves formed, but qualitatively shows the effect of the Stokes shift and refractive index on them.

It is clear that for a homogeneous medium, when the threshold is exceeded and the pumping intensity is uniform in cross-section, the period of the resulting surface wave will be constant. This is clearly seen in Fig. 1, *c*, which is obtained at a uniform cross-section intensity. But since the refractive index depends on the intensity of the interacting waves, then in the event of unevenness of their intensity in the transverse direction, the period of surface waves will track it. This applies equally to radial scattering within the medium, changing the radial wave period of $\lambda_{\text{st}}/n_{\text{st}}$, and to scattering in the surface layer, changing the period of surface waves.

Since in our case we are interested in Stokes waves scattering along the surface, the increase in the refractive index for them, depending on the intensity of pumping, leads to a decrease in the spatial period of their beats with the pump wave, or the period of the surface waves formed.

As a rule, the distribution of pumping intensity along the beam radius is uneven, and with increasing radius, the intensity decreases, which should lead to an increase in the wave period.

The formation of a large number of periods of surface waves covering almost the entire area of laser exposure (Fig. 1, *c*), despite the fact that due to scattering in the radial direction, a weakening of Stokes waves occurs, is possible only by amplifying the Stokes wave (forced phonon-polariton radial scattering).

If there is an angle β between the normal to the surface and the beam incident on it, then the interaction of the pump wave with the Stokes wave propagating along the surface will be determined by the effective pump wave length, as $\lambda_p^{ef} = \lambda_p \cos\beta$. In this case, the formula for the period of the bands will be recorded as

$$\lambda_{be} = \frac{\lambda_p \lambda_{\mathrm{st}}}{\dfrac{n_p \lambda_{\mathrm{st}}}{\cos\beta} - n_{s\lambda_p}},$$

and, depending on the direction of propagation along the surface of the Stokes wave, the wave period will change in a positive or negative direction.

In the case of focusing radiation into a medium, the angle of inclination of the rays to the surface in a focused wave with a spherical front is determined from the condition $\cos\alpha = R/r$, where R — depth of focus under the surface, and r — radius at which the beam reaches the surface, which, with a large numerical aperture ($\mathrm{NA} = 2r/f$) and an intensity independent of the radius, should lead to a decrease in the period of the rings with an increase in their radius.

But usually in experiments, the beams are directed normally to the surface, and the focusing is carried out with sufficiently long-focus lenses, which makes the angle β quite small.

On the other hand, the decrease in the refractive index due to the attenuation of the pump wave with the removal from the axis of the focused beam (usually the Gaussian distribution) has a much stronger effect on the increase in the period of the formed surface waves, which we see in Fig. 3 from the study [7]. This study has investigated the formation of concentric rings on the surface of the crater in SiO_2, formed when exposed to it by a laser pulse with a duration of 12 ns at a wavelength of 1064 nm. The laser emitted a beam with a Gaussian profile in TEM_{00} mode. Judging by the profile of the surface of the crater, presented in the study [4], then its formation, basically, occurred not due to ablation, but due to the plastic deformation of the surface with the squeezing of the medium in the form of a rim on its edges, which affected the width of the edge rings.

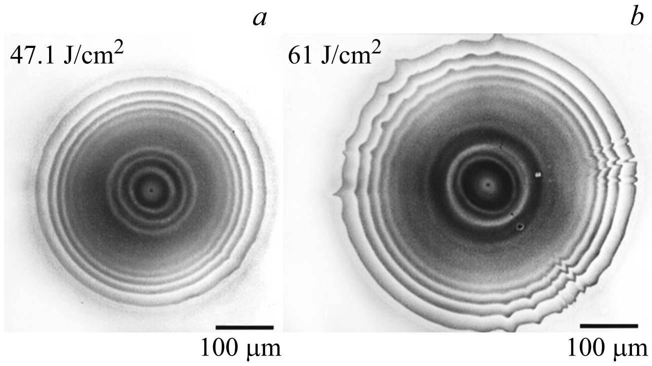

Figure 3. Concentric rings formed on the surface of the slopes of the crater in SiO_2 when focusing a laser beam with a Gaussian intensity distribution in the mode TEM_{00}. $\tau = 12$ ns, $\lambda = 1.064\,\mu$m.

Regular concentric circles were observed with a change in laser energy from 19.6 J/cm^2 to 61 J/cm^2. The depth of the damage spots in the center, depending on the intensity of pumping, ranged from 38.1 to 79.8 nm. Hence, the angle of inclination of the slopes of the crater in Fig. 3 is several hundredths of a degree, so the main contribution to the change in the period of the rings with radius formed on them is the exponential weakening (Gaussian distribution) of the intensity of the pump wave with radius, which reduces the nonlinear refractive index. And since the nature of the change in intensity from the radius does not change with a change in the pumping energy, the dependence of the period of the rings on the radius with the same inclination of the slopes of the crater remains constant, only the zone of their formation with the increase in energy shifts towards large radii (Fig. 3, *a, b*), and in the central region rings with a different period appear. Perhaps this is due to the excitation of Stokes waves with different frequency shift, which gives two ring systems with different periods.

In the study [7] it is suggested that the rings in Fig. 3 are the result of interference of the pump wave in the interval between the shock wave propagating deep into the medium and the surface of the sample. In this case, we would have an interference pattern similar to Newton's rings with their period decreasing towards the edge, rather than increasing as in these figures. This is due to the fact that in the center the depth of penetration of the shock wave deep into the medium and its amplitude are maximum. That is, we would have to observe a pattern of interference approximately corresponding to that formed when the test pulses of the flying liquid shell from the surface of the sample are translucent under the influence of femtosecond pulses [10]. Since the interference pattern is dynamic, the bands shifting during the pulse action, when the shock wave is shifted at a maximum of $\sim 100\,\mu$m, would smooth out the result of interference.

In Fig. 1, *c*, the focusing was done by a long-focus lens, and the intensity in the pump plane was almost constant, which led to an almost constant pitch of the concentric rings. The rings on Fig. 3, formed on the surface of the crater during acute focusing, increase their period due to a continuous weakening of the intensity with radius.

Thus, the rings on the surface are the result of the orthogonal interaction of the pump wave with scattered Stokes waves from the boundaries of the central crater (in the case of Fig. 3 it is the crater $\varnothing \sim 5\,\mu$m) in the surface layer, in some time, as the craters themselves — this is the result of the development of stimulated radial scattering germinating within the medium from seed heterogeneity to surface.

In Fig. 3, *b* it can be seen that concentric rings are not smooth, obeying the harmonic law, but have a sharp edge on the inside. This is explained by the fact that the forces acting on the surface during radial scattering are directed not along the normal to the surface, but at an angle $\sim 45^\circ$ to it in the direction of their center, which leads to the asymmetry of the wave crests, making their leading edge directed inward steeper.

The source of Stokes waves, in this case, is a central microcrater with a diameter of $\sim 5\,\mu$m, the size of which grows with increasing pumping, which is a continuation of the channel formed by the germination of radial scattering waves to the surface. With an increase in the intensity of pumping, the threshold for the development of radial scattering on smaller inhomogeneities in the central region of the focus spot is exceeded [4]. New, offset from the axis craters (centers of scattering) appear. At high magnification near the center of the crater, many tiny pits were observed [7]. Scattering from them when superimposed on scattering from the central crater leads to interference wave-like distortions of the outer rings. The influence of additional microcraters to the right of the central one, distorting the edges of the rings, is visible in Fig. 3, *b*, which once again indicates the mechanism of their formation: the interaction of the pump wave with Stokes scattering from the walls of various craters near the axis, which are the result of the impact of stimulated radial scattering.

As for the wave period on a flat surface, it should depend on the magnitude of the Stokes displacement, and the smaller it is, the more times the Stokes waves and the pump wave will coincide their phases on a flat surface. Naturally, it is necessary to take into account the refractive index for the Stokes wave, which in the case of a resonant interaction does not coincide with n_1.

The number of periods of surface waves formed and their blurring, except for the surface of the irradiation zone and the intensity of the interacting waves, depends on their spectral width, and with an increase in width over large periods, it is possible to overlap the maxima, leading to the erosion of the waves.

But the number of periods of the waves formed also depends on the curvature of the boundaries from which the Stokes waves scatter at the same amplification coefficient, since the smaller their curvature $(1/R)$, the smaller their radial weakening (smaller scattering angle), which contributes to an increase in the number of periods of the resulting waves.

By converting the beat formula to the form,

$$n_{\text{st}} = \lambda_{\text{st}} \left(\frac{n_p}{\lambda_p} - \frac{1}{\lambda_{be}} \right)$$

it is possible to estimate the refractive index that would explain the observed period of the rings. Using this formula for Ge $(\Delta\nu = 300\,\text{cm}^{-1})$ in Fig. 1, *c*, with a ring period of $\sim 25\,\mu$m we get $n_{\text{st}} \approx 5.2$, which exceeds the refractive index for germanium $n = 4$. At the same time, the pumping intensity was $I = 4 \cdot 10^8\,\text{W/cm}^2$. For rings in YAG $(\Delta\nu = 370\,\text{cm}^{-1})$ in Fig. 1, *a*, the period of which is $\sim 10\,\mu$m, the refractive index is $n_{\text{st}} \approx 11$ at pumping intensity $I = 5 \cdot 10^9\,\text{W/cm}^2$. Similar estimates for amorphous SiO, according to Fig. 2, *b* give us for the period $\sim 6.7\,\mu$m the refractive index $n_{\text{st}} \approx 7.6$. Such a high refractive index indicates an anomalous dispersion in the region of the resonant interaction of the Stokes wave with the pump wave. But this is the average refractive index for a given period, which abnormally increases at the wave maxima and decreases at the minima.

3. Surface fractures in the form of intersecting straight lines

If the scattering does not occur on a point defect, but on a linear inhomogeneity — on a surface scratch or an extended edge dislocation located close to the surface, then we have radial scattering from each point of inhomogeneity transformed into bidirectional scattering across the direction of inhomogeneity, and forms in the medium not conical converging on the axis acoustic waves, but wedge-shaped waves along the inhomogeneity converging at the same angle $(\sim 45^\circ)$, and forming longitudinal channels of penetration on the surface. The development of stimulated scattering at edge dislocations is facilitated by the fact that elastic dislocation fields lead to an increased concentration near its axis of point defects with their reduced binding energy, significantly increasing scattering. The edge dislocations themselves are more often formed along the planes of adhesion, along which the binding energy of atoms is weakened. As a result, fractures are sometimes formed on the surface in the form of straight intersecting lines directed along the planes of adhesion (Fig. 4, *a*), observed on the surface of the YAG crystal when pumped with a nanosecond pulse with a wide spectrum [4], and on the surface of the crystal Ge (Fig. 4, *b*) from the study [6]. Similar straight intersecting lines were observed on the input surface of sapphire when exposed to it by a powerful nanosecond pulse in long-standing study [11].

Thus, radial waves formed at different points on narrow transverse inhomogeneities of the medium are transformed

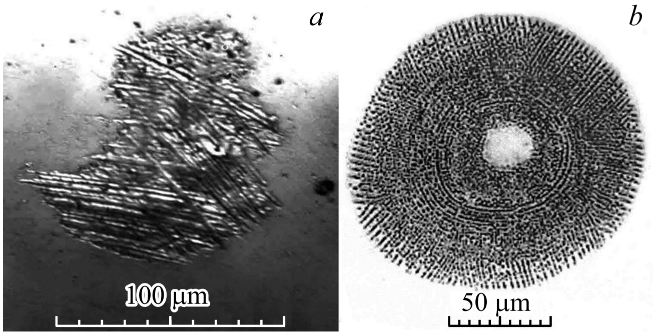

Figure 4. *a* — fracture spot on the surface of the YAG crystal with intersecting surface punching lines located along the adhesion planes. $\lambda = 0.92\,\mu$m, $I \sim 5 \cdot 10^9$ W/cm^2, *b* — microdestructions on the surface Ge in the form of concentric rings and intersecting straight lines directed at the angles of adhesion [110]. $\lambda = 10.6\,\mu$m, $I \sim 4 \cdot 10^7$ W/cm^2.

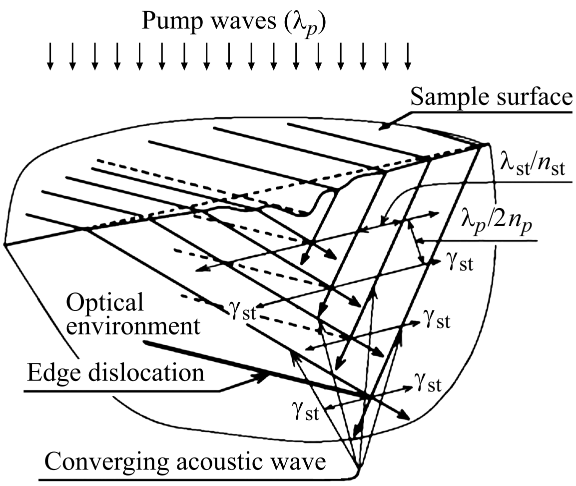

Figure 5. A diagram explaining the mechanism of formation of longitudinal grooves on the surface along the edge dislocations present in the environment due to the wedge-shaped convergence of shock waves on them.

into periodic waves of transverse scattering. A simplified diagram of the formation of longitudinal surface grooves during their development from edge dislocations is presented in Fig. 5. The figure shows the formation of convergent longitudinal acoustic (shock) waves developing from the interaction of the pump wave (λp) and the Stokes wave ($\lambda_{st}, \gamma_{st}$) of Raman scattering on the longitudinal heterogeneity (edge dislocation), the impact of which leads to the formation of longitudinal grooves on the surface.

The very process of pushing, which takes place when a periodic sequence of shock waves is exposed to the medium, occurs due to the intensive generation of point defects in their field, and the displacement of the resulting interstitial atoms under the action of shock waves deep into the medium. At the same time, there is a process of counter movement of vacancies to the surface with the formation of depressions. The presence of parallel edge dislocations in the medium manifests itself on the surface in the form of parallel lines. The dislocations running at other angles along other planes of adhesion form intersecting lines.

4. Manifestation in surface layer of radial scattering waves

A micrograph of the destruction (Fig. 4, *b*) on the surface Ge from the study [6] was obtained by irradiating the sample with a wide weakly convergent beam of a microsecond laser at CO_2 with an intensity of $I \sim 4 \cdot 10^7$ W/cm^2. It can be seen that around the central zone there is an area filled with concentric rings, and bordering them with a region of intersecting lines oriented in the direction [110]. The period of these rings and intersecting lines is significantly less than the pump wavelength, and they cannot be the result of the beating of interacting waves. It remains to be assumed that they are the result of the manifestation in the surface layer of radial scattering waves within the medium, which have a period of λ_{st}/n_{st}. Since the interaction of Stokes waves scattered in the radial direction with the pump wave in the medium occurs at the angles of synchronism with the period λ_{st}/n_{st}, and not with the period of their beats, the threshold intensity for radial scattering will be significantly lower than for the excitation of surface waves. This indirectly confirms that the formation of local destruction on the surface Ge (Fig. 4, *b* from the study [6]) occurs when the sample is irradiated with a wide weakly converging beam of a microsecond CO_2 laser with intensity of $I \sim 4 \cdot 10^7$ W/cm^2, and the formation of significant surface destruction with the formation of periodic surface concentric rings (Fig. 1, *c*) under the same focusing conditions occurs at the intensity of $I \sim 4 \cdot 10^8$ W/cm^2.

Usually, we do not notice the manifestation of radial scattering on the surface due to the small pump wavelength, but in this case, at $\lambda_p = 10.6\,\mu$m, their period becomes noticeable. It smoothly varies from $\sim 2.25\,\mu$m for rings located at a distance of $\sim 25\,\mu$m from center, to $\sim 3.0\,\mu$m for intersecting lines at the edges in $\sim 65\,\mu$m from center, which most likely indicates a decrease in the refractive index at a distance from the center, caused by attenuation with the radius of intensity of the Stokes wave. The average refractive index between the maxima of the generated waves (Δr) can be determined by knowing the length of the Stokes wave $n_{st} = \lambda_{st}/\Delta r$. If we assume that the wavelength of Stokes scattering for Ge at Raman shift $\sim 300\,\text{cm}^{-1}$ is equal to $\lambda_{st} \sim 15.5\,\mu$m, then refractive index when the distance from the center changes from $25\,\mu$m to $65\,\mu$m, estimated by this formula, varies from ~ 7 to ~ 5, while the normal value $n = 4$.

If the period of excited transverse waves changes due to a change in the refractive index with distance from the center, then the excitation of radial waves at adjacent dislocations at distances multiple of the period of these waves becomes impossible, and their development occurs on other dislocations, the distance between which is more consistent with the period of waves. Because of this, parallel waves, when they germinate from the center, develop not in the form of continuous lines, but in the form of discontinuous (Fig. 4, *b*), which abruptly adapt to new conditions. This, to a large extent, applies to concentric rings, which are easier to develop on defects in the crystal lattice (dislocations). Therefore, we see that often the rings do not form continuous circles, but consist of small straight segments.

This picture can be considered a visual manifestation of stimulated radial scattering near the surface, which becomes observable due to the large wavelength of the pump radiation.

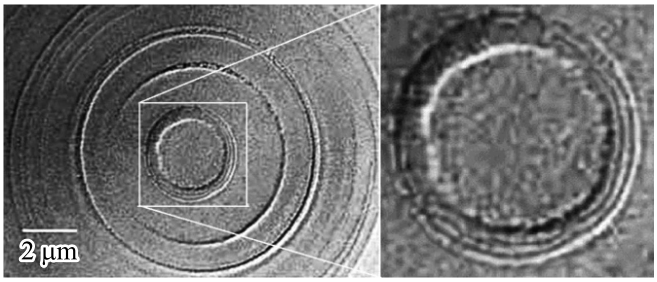

Figure 6. The rings on the surface of the crater in soda-lime glass, formed when exposed to momentum $\tau = 20$ ns, $\lambda = 1.064\,\mu$m, $I = 5.8\,\mathrm{W/cm^2}$. On the right there is an enlarged center part.

5. Formation of a system of narrow concentric rings on the surface of the resulting crater

The formation of rings on the surface of the crater is possible if, during the impact of the pump pulse, radial scattering develops, which is an intense source of radial Stokes waves. As the crater deepens, with nanosecond pulses, the conditions for phase synchronism between radial waves and the pump wave change, and annular maxima are formed on the concave surface of the crater with its own period. The period of the radial rings will depend on the steepness of the slopes of the crater, on which the phasing of interacting waves occurs. At the same time, the conditions for the formation of radial rings on its surface are constantly changing, adjusting the period of the resulting waves to them.

At those points where the Stokes waves running from the axis of radial waves tangentially to the curved surface are in the phase with the pump wave, resonant molecular oscillations (phonons) are excited with the transfer of scattering difference pulses to the molecules. It is at these points of the circle, symmetrically separated from the axis, due to plastic deformation and ablation, the formation of narrow ring zones occurs on the surface.

The most visually similar structures are presented in Fig. 6, which were observed on the surface of soda-lime glass when exposed to it 20 ns pulses on $\lambda = 1.064\,\mu$m in [12]. In this case, the surface of the resulting crater was close to spherical, and due to the continuously changing slope of its walls, the thin rings forming on its surface did not form periodic structures, but formed groups of several narrow concentric rings. The formation of narrow rings in this figure, the narrowest of which are ≤ 100 nm, with a minimum distance between them ~ 150 nm, which is significantly less than the Stokes wavelength, cannot be explained by interference, but is explained as a manifestation of radial scattering waves. The same narrow rings, the distance between which in groups is less than the wavelength, with a slightly different distribution in radius, were observed on the surface of fused quartz. At the same time, in the figures with a larger coverage area at large radii, double and built lines are observed, which can be explained as a manifestation of the radial scattering wave period, which increases with a decrease in the intensity of the Stokes wave at a distance from the source due to a decrease in the refractive index. The minimum period of rings observed in the insertion of Fig. 6 from the central region is $\Delta r \approx 150$ nm, which, given that $\Delta r = \lambda_{st}/n_{st}$, once again indicates an abnormally high refractive index for radial Stokes waves.

6. Conclusion

In the paper, based on the analysis of wave-like destruction on the input surfaces of various optical media formed under the action of powerful laser radiation, a mechanism is proposed to explain their formation. It is shown that the formation of waves occurs not due to plasmon-polariton scattering, as in the case of femtosecond pulses, but due to the phonon-polariton, which underlies the stimulated radial scattering, which makes the period of surface waves dependent on the Stokes displacement, and the anomalous dependence of the refractive index on the intensity of the interacting waves contributes to their period. Phase modulation of orthogonally interacting Stokes waves of radial scattering in the surface layer and pump waves leads to an uneven effect on the surface layer, causing its deformation in the form of concentric surface waves, in the case of axisymmetric scattering, and in the case of asymmetric scattering, periodic waves track the boundaries of scattering inhomogeneity. The same mechanism, in the presence of transverse edge dislocations in the medium, leads to the formation of narrow transverse grooves on the surface, elongated along the planes of adhesion of crystals. The small width of the bands and rings formed in this case on the surface, the much smaller wavelength of the acting waves, as well as the distance between them is also less than the wavelength, allows us to conclude that we observe the manifestation of stimulated radial scattering in the surface layer, which occurs with a significant increase in the refractive index due to the resonant interaction of waves (anomalous dispersion). It is noted that in the case of large wavelengths (section 4) or a curved surface (section 5), it is possible to observe surface waves with a period of radial scattering waves.

Naturally, such structures could not be formed in the case of reflow of the surface, since surface tension forces would smooth the surface.

Conflict of interest

The author declares that he has no conflict of interest.

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