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INVESTIGATION OF LASER-INDUCED DEPTH DAMAGE AND SCATTERING OF LIGHT IN CRYSTALS AND GLASSES

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Laser-induced depth damage to sapphire crystals and to various glasses is investigated. The influence of self-focusing on the laser damage process is studied. The conditions under which self-focusing influences the damage are clarified. The influence of various impurities on the optical endurance of ruby laser crystals is determined. The damage mechanism is initiated by various types of absorbing inclusions and defects; a correlation is found between the light scattering and the damage threshold. A new criterion based on light scattering is introduced for the purity of transparent dielectrics.

1. SURVEY OF RESEARCH ON LASER DAMAGE TO TRANSPARENT DIELECTRICS

Damage to transparent dielectrics was observed immediately after the advent of lasers with giant emission pulses [1]. This was followed also by investigation of damage produced by millisecond laser pulses [2-4]. The mechanisms responsible for the damage were not identified in the cited references. It must be noted, however, that the adverse effect of metallic platinum particles on the damage threshold of laser glasses was elucidated in [3].

Hercher was the first to observe the filamentary character of the damage to glasses by giant ruby laser pulses focused into the interior of the samples [5]. A similar damage picture was observed in various types of transparent dielectrics by Ashkinadze et al. [6]. Budin and Raffy attempted to explain the damage mechanism by investigating the time evolution of the depth damage to glasses by ruby laser emission of 30-nsec duration [7]. It was established as a result that the damage has a filamentary character and develops along the laser beam at a velocity exceeding $5 \cdot 10^7$ cm/sec. The dimension of the produced filament turned out to be smaller by two orders of magnitude than the dimension of the focal spot, and this, in the authors' opinion, served as direct evidence of the presence of self-

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focusing in solids. The filamentary pattern of the damage was later explained on the basis of the theory of moving foci [8].

The position of the laser-radiation focusing lens relative to the boundaries of the transparent medium influences the threshold and the character of the produced damage. A distinction is made in this connection between three types of damage to transparent dielectrics.

1. Damage to the Entrance Face. It occurs when the beam is focused on the entrance surface and is always accompanied by a bright spark.*

2. Damage to the Exit Face. It occurs when the laser beam is focused on the rear surface of the sample and it not always accompanied by a spark. The threshold is smaller by a factor of 1.5-2 than in the first case.

3. Depth Damage. This type of damage is most frequently investigated and takes place as a rule when the radiation is focused into the interior of the sample; it is accompanied by high-temperature emission. The thresholds P_s and P_d of the surface and depth damage differ substantially in most cases: $P_s \ll P_d$.

These three types of damage were first observed by Guiliano [1]. He attempted to explain the experimental results on the basis of the mechanism of generation of acoustic phonons via stimulated Mandel'shtam—Brillouin scattering (SMBS), followed by their damping and conversion of their eneergy into heat. However, as shown in [10], this mechanism is not decisive and cannot explain the observed strong difference between the thresholds of the depth and surface damage.

It was suggested in [11] that the surface is less resistant because of the presence of a strongly absorbing surface layer. This question is the subject of [12], in which a study was made of the dependence of the threshold of surface damage on the duration of the laser pulse. It revealed distinctly for the first time the role played in the damage mechanism by the absorbing defects in the surface layer. These defects are estimated to measure approximately 3-5 μm and to have an absorption coefficient $\alpha \sim 10^2$ cm^{-1}.

Bonch-Bruevich et al. [13] attempted to investigate the optical properties of the surface layer. They observed with the aid of infrared radiation the kinetics of the heating and cooling of the surface layer of optical glass under the influence of the emission of a ruby laser in the usual lasing regimes at intensities below the surface-damage threshold. The layer thickness was estimated at 10-20 μm and the coefficient of linear absorption of light by this layer was $\simeq 1$ cm^{-1}. The question whether the entire layer absorbs uniformly or in small volumes, as shown in [12], remained open. It is natural to assume that the surface layer in glasses, just as in sapphire crystals [12], is not a continuous absorbing layer, but but consists of individual absorbing defects. Then the effective absorption ~ 1 cm^{-1} can be explained if the volume concentration of the defects having $\alpha \sim 10^2$ cm^{-1} amounts to 1%.

Fersman and Khazov [14, 15] have proposed that the surface damage is due to interaction between the plasma produced in the optical breakdown and the surface, and explained the difference between the damage to the entrance and exit faces as being due to the fact that the plasma tends to grow in the direction opposite to that of the laser beam. On the basis of this model one can expect the rear face to be more damaged than the front face, since the plasma on its surface tends to press against it, whereas the plasma on the front face tends to break away from it. The authors of [14] observed a compression wave propagating into the interior of the sample from the rear surface and offering evidence, in their opinion, of interaction between the plasma and the surface. The compression waves produced at the points of damage were observed also by other workers using both shadow procedures [16] and holography [17]. Although the proposed model is indeed capable of explaining the difference in the damage to the entrance and exit faces, it is not experimentally confirmed, as shown in [19], where it was demonstrated that the plasma is more readily the consequence rather than the cause of the surface damage. What is universally accepted is the fact that the threshold of the laser-induced surface damage to transparent dielectrics depends on the state of the surface. Extraneous impurities in the surface layer can cause damage at light intensities much lower than those that can be withstood by pure surfaces. The quality of the surface finish also influences the optical endurance [18, 11]. Numerous attempts were made

*We note that Giuliano [9] observed surface damage without spark formation, but only in a very narrow interval of the laser-radiation intensities near the threshold.

of chemically strengthening the surfaces. Some procedures led to an increase of the endurance, but frequently this effect was temporary. Davit [19] has demonstrated that in action on surfaces of various glasses in which the hydroxyl groups OH are replaced by CH_3 the damage threshold is raised from 40 to 60 J/cm^2 for 30-nsec laser pulses and when the OH is replaced by F the threshold increases to 110 J/cm^2. This value of the threshold, however, is preserved for several minutes, after which it decreases rapidly to 40 J/cm^2. Similar results were obtained also in [20]. The authors of [21] have likewise not observed any substantial change in the surface-damage threshold of ruby subjected to various cleaning procedures. Only by using ion polishing [22] and thermal evaporation [23] of the ruby surfaces was it possible to substantially increase their optical endurance.

The process of laser damage to transparent materials can be arbitrarily broken up into two stages. The first consists of laser-energy absorption, which can be due to the presence of impurities or can be an intrinsic property of the pure material. During the second stage, the absorbed energy leads to a local temperature rise and to an irreversible change of the material. The conditions under which the second stage becomes insignificant in the analysis of the damage were considered by many workers. Conners and Thompson [24] formulated the problem of laser damage from the point of view of the dynamic theory of thermoelasticity, with the aim of determining the amount of energy that must be absorbed from the laser beam in order to produce damage. A detailed analysis of this question was carried out by Sharma and Rieckhoff [25]. They investigated the influence of the duration of the laser pulse and of the focal length of the focusing lens on the distribution of the stresses in the medium. As a result they obtained a criterion for laser damage of the material, namely, the effective light absorption coefficient in the focal volume near the damage threshold should be $\alpha \simeq 50$ cm^{-1} for borosilicate glass for a ruby laser pulse of duration 30 nsec with a peak power density of $3 \cdot 10^9$ W/cm^2. The analysis of the dynamics of the damage then becomes inessential. This value of α cannot be accounted for by linear absorption of light by a uniformly absorbing medium. Therefore the mechanisms of nonlinear absorption should play a substantial role in the damage of pure materials.

On the basis of the foregoing, various mechanisms of nonlinear absorption of light capable of leading to damage of pure dielectrics were considered. Kroll [26] considered the problem of photoelastic instability in SMBS in quartz and sapphire under the influence of powerful laser radiation. An estimate of the effective absorption coefficient for this process yielded a value $\alpha \simeq 0.1$ cm^{-1}, much less than the value required ($\sim 10^2$ cm^{-1}) for mechanical damage. Ritus and Manenkov [10] investigated the thresholds of SMBS and of depth damage in glass and in fused and crystalline quartz. The fact that the damage in many substances is observed below the SMBS threshold indicates that the SMBS cannot be responsible for the damage.

An effective absorption coefficient $\alpha \sim 10^2$ cm^{-1} can be achieved in a pure dielectric via absorption by free electrons in the conduction band, if their concentration exceeds 10^{18} cm^{-3}. Such high concentrations can result from impact ionization (the electron avalanche mechanism) in the material matrix following the passage of a laser pulse, or on account of multiphoton absorption of the incident radiation [27]. We shall dwell in detail on the electron avalanche mechanism, since in many papers it was precisely this mechanism which was assumed to be responsible for the damage to transparent dielectrics in the case of nanosecond laser pulses.

The electron avalanche mechanism, according to present-day notions, evolves in a pure dielectric in the following manner: in a region with a strong local electric field, the initial electrons are produced via multiphoton ionization of the atoms and absorb the light quanta in collisions with the atoms or ions. After acquiring an energy somewhat higher than the ionization potential, each electron ionizes an atom with high probability. As a result of this process, one electron generates two electrons with lower energy, which initiate the entire cycle anew. The presence of initial free electrons is sometimes attributed to the presence of a small number of impurities in the medium.

Attempts to calculate the electron avalanche mechanism in pure dielectrics, as applied to optical fields, were made by many workers [3, 25, 28, 29]. Zverev et al. have extended Fröhlich's theory [30] to include the case of hf electric fields, assuming that the laser frequency is much higher than the frequency of the electron–phonon collisions. An estimate of the critical field for damage in sapphire yielded the value that exceeded by more than one order of magnitude the experimentally measured one. A similar extension of Fröhlich's theory to include damage to alkali-halide crystals was reported in [31]. In this case, too, the calculated fields were much stronger than the measured ones.

The Fröhlich theory [30-33], developed for breakdown of dielectrics in constant electric fields, is based on the assumption that the electrons lose energy when they interact with longitudinal optical phonons (a quantum-mechanical calculation of such an interaction was presented in [34]). The breakdown fields calculated in this case turned out to be much higher than those observed in experiment.

Seitz [35] has indicated that electrons in ionic crystals can interact not only with longitudinal optical phonons, but also with acoustic phonons. The latter interaction can predominate at electron energies exceeding the energy of the optical phonon, thus leading to an increase in the effective frequency of the electron—phonon collisions and to a decrease in the values of the critical fields. In this case, however, the calculated breakdown fields for NaCl turned out to be much higher than those observed in experiment. Seitz attributed this discrepancy to the fact that the avalanche can be the process not for the average electron, and that the fluctuations of the electron energy can substantially lower the damage threshold. He has also proposed a "40 generations" criterion, which explains qualitatively the dependence of the threshold fields on the sample thickness [36, 37].

Molchanov [29] considered an electron avalanche in dielectrics with allowance for the scattering of the electrons by acoustic phonons. Using the "40 generations" criterion he obtained agreement between the calculated threshold field of the optical breakdown and the experimental value for sapphire crystals. This agreement is apparently accidental, inasmuch as no account was taken in the calculation of the intraband scattering of the electrons by acoustic phonons; this scattering, as shown in [38], is quite substantial and greatly increases the values of the critical fields. In [38] there were proposed also other variants of the electron avalanche mechanism with participation of multiphoton processes, but all the calculations yield threshold fields considerably exceeding the experimentally observed values. A similar result is obtained also when multiphoton absorption in a pure dielectric is considered [27].

As a result, attempts were made to explain the experimental data on the damage to transparent dielectrics by invoking such mechanisms as self-focusing and absorption of light by foreign inclusions. The substantial influence of absorbing inhomogeneities on the optical breakdown threshold follows from direct experiments [12, 39, 40]. It is shown in [41] that the breakdown threshold of a liquid dielectric increases monotonically to the extent that the liquid is rid of microscopic solid particles. This raises the question of the mechanisms that produce laser damage to transparent dielectrics containing absorbing inclusions. The simplest mechanism, which is effective in the case of relatively large inhomogeneities, is local melting and formation of cracks as a result of thermoelastic stresses. A linear theory of the thermal damage mechanism was developed (without allowance for the dependence of the parameters of the medium on the temperature); this theory is described most completely in [39]. However, the conclusions of this theory (and in particular concerning the dimensions of the dangerous inclusions) are not sufficiently well founded, inasmuch as high temperatures ($\sim 10^4$ K) are reached in the laser damage process, so that it is impossible to disregard the temperature dependence of the parameters of the medium. In addition, the linear theory also fails to provide a qualitative explanation of the well-known fact of the clearly pronounced threshold character of the damage, which is accompanied by high-temperature glow (spark).

A nonlinear theory of thermal damage with allowance for the temperature dependence of the parameters of the medium was developed in [42] and is described in detail below. In contrast to the linear theory, the nonlinear theory accounts well for the experimental data on laser damage (the threshold character of the damage and the high-temperature emission as well as the threshold intensities of the radiation). If the absorbing defects are too small to cause microdamage in the host matrix, they can nevertheless initiate absorption by the initially transparent medium that surrounds the defects.

Impurities in transparent dielectrics can also strongly influence the multiphoton ionization of the medium in the field of an intense light wave [27]. In this case the ionization of a solid is possible via intermediate states, whose role may be played by local levels of impurities and crystal lattice defects if their concentration is not very small. The probability of multiphoton ionization may then increase by several orders of magnitude, leading to a substantial decrease of the incident radiation intensity necessary for ionization and subsequent damage. Hellwarth [43] has shown that owing to the strong electron-phonon coupling in the sapphire lattice no electron avalanche can develop in it. He has therefore proposed multiphoton and cascade ionization of the impurities as one of the possible mechanisms. Estimates on the basis of this mechanism have shown that allowance for the presence of impurities in a real dielectric turns out to be substantial and can explain the observed values of the threshold intensities.

The question of the damage to transparent dielectrics is closely connected with the question of self-focusing: damage can proceed through a stage of self-focusing, and the intensity of the radiation passing through the medium is then substantially increased. The damage threshold in this case is determined not by the optical endurance of the material itself, but by the self-focusing threshold. Budin and Raffy [7] were the first to attribute the filamentary damage in glasses to self-focusing of the radiation. In later studies, Zverev et al. [44, 45] observed a similar character of the damage in sapphire and in K-8 glass. The filamentary damage picture was explained on the basis of the theory of moving foci [8], the velocity of which $\sim 10^8$ cm/sec was measured in [45]. A standing pattern of self-focusing points in glass and a corresponding damage pattern were observed by Lipatov et al. [46] in the case of rectangular radiation pulses. The self-focusing effect limits the maximum radiation intensity attainable in high-power laser systems used for laser-induced thermal poisoning [47]. A filamentary picture of the damage is observed in this case in the active laser elements of the amplifier output stages; this picture is due to self-focusing of a parallel laser beam (in contrast to [7, 8, 44, 45], where self-focusing of prefocused beams was observed). Even this small list of studies shows that the self-focusing effect exerts a strong influence on the damage process and demands special analysis in investigations of laser damage to transparent materials.

It has been recently reported that in the investigation of damage to certain crystals and glasses it was possible to avoid the influence of self-focusing [48-50] and to obtain good experimental agreement with the damage theory based on the mechanism of the electron avalanche that develops in pure material. It will be shown below, however, that this conclusion cannot be regarded as well founded.

Summarizing the results of the numerous early investigations (prior to 1970) aimed at determining the optical-damage mechanisms, it must be indicated that many circumstances have prevented comparison of the experimental results of different authors and their adequate interpretation on the basis of the existing theories. Among these circumstances are the uncertainty of the mode composition of the radiation [51], the lack of a clear-cut criterion of optical purity of the investigated materials [21], the undesirable influence of self-focusing, which hinders the determination of the radiation density in the damage region [52], and the absence of sufficient theoretical grounds for the interpretation of the results. The need for performing experiments with single-mode lasers became obvious following the publication of [53, 54], where it was shown that the damage mechanisms can be different when single-mode and multi-mode lasers are used.

Notwithstanding these difficulties, important results were obtained. First, it was established that in most cases the laser damage was due to absorbing inclusions, so that their elimination has become the task of technological control of the material preparation conditions [55, 56]. Second, it was found that the self-focusing effect leads to a considerable increase of the radiation intensity inside the medium, and that the threshold of the depth damage is in fact the self-focusing threshold. To investigate the mechanism of laser damage to a pure dielectric it is therefore necessary to eliminate the influence of absorbing defects and of self-focusing.

The first research investigations, in our opinion, were successfully performed in [57, 48-50] in the studies of damage to certain crystals and glasses. The authors of the cited papers stated that they found a simple procedure for revealing the cases of intrinsic damage to transparent dielectrics and damage initiated by inclusions, and that they had excluded the influence of self-focusing.

To investigate the intrinsic damage to dielectrics, Yablonovitch [57] selected as the most suitable objects alkali-halide crystals, whose damage characteristics were investigated in constant electric fields [58]. He found the correlation between the critical fields for optical breakdown at λ = 10.6 μm and for a dc electric breakdown.* It was concluded on this basis that the only mechanism responsible for laser damage is electron avalanche in the pure material. This conclusion, in our opinion, is not fully justified for the following reasons: 1) The employed laser did not generate a single frequency, and the radiation pulse was subject to nonreproducible time modulation; 2) the investigated samples were of low optical quality, as evidenced by the presence of damage at the absorbing inclusions; and 3) it is incorrect to compare the thresholds of the breakdown in constant electric fields [58] and laser breakdown, in view of the presence of hard-to-control factors in investigations of breakdown in constant fields (the influence of the electrodes, the purity of the materials, etc. [36, 37]).

The ideas advanced in [57] are further developed in [48, 49, 60] in which, in the authors' opinion, it was shown that the decisive mechanism in the damage to alkali-halide crystals by 1.06- and 0.69-μm laser radiation is also the electron avalanche. This conclusion was based, just as in [57], on the equality of the measured breakdown thresholds at optical frequencies as well as in a dc field, and on the absence of influence of self-focusing on the process of laser damage under their experimental conditions. However, the conclusion drawn in [48, 49] that the decisive role is played by the electron avalanche mechanism cannot be regarded as proved, for the same reasons as in the analysis of [57] (i.e., poor optical quality of the investigated materials and incorrect comparison of the data on the laser damage and breakdown in constant electric fields). As for the statement that self-focusing exerts no influence on the damage process in the case of sharp preliminary focusing of the radiation, it is likewise subject to doubt for the following reasons. The authors of [48, 49] believe that the self-focusing effect is determined by the total power of the radiation entering the sample and does not depend on the size of the beam. However, as shown in [61], when the radius of the focal spot is decreased from 140 to 20 μm the total input power at which damage due to self-focusing takes place is decreased by a factor of 2.5. This points to a certain dependence of the self-focusing on the beam size [62], and calls for a detailed investigation of this phenomenon in the case of sharp focusing of the laser radiation, something not done by the authors of [48, 49]. In addition, the results of the experimental verification that self-focusing exerts no influence on the damage process [63] are not quite convincing. Bass and Fradin have found that the threshold power of the laser radiation P_d varies with the focal length F of the focusing lens like $P_d \sim F$. Such a dependence was taken to be proof of the absence of any influence of self-focusing, whereas absence of this influence calls for a relation $P_d \sim F^2$. Thus, the results of the experimental verification cited in [48, 49, 63] do not give sufficient grounds to regard the absence of influence of self-focusing on the laser-damage process as proved.

The independence of the damage threshold of the laser frequency, which is assumed by the authors of [48, 49, 57] to be one of the arguments in favor of the decisive role of the electron avalanche mechanism, can be easily explained on the basis of the thermal damage mechanism, inasmuch as the absorption of light by inclusions and defects does not have a resonant character.

One of the proofs, in the opinion of the authors of [64, 65], of the decisive influence of the electron avalanche on the damage process is the statistical character of the laser damage (both in depth and on the surface), which was investigated in various materials, predominently nonlinear crystals. It was found that the probability of damage at a certain value of the electric field E is proportional to $\exp(-k/E)$, where k is a certain constant determined by the properties of the given material. The observed statistical character of the damage cannot be attributed to fluctuations of the laser power, and is treated by the authors on the basis of the model of the "fortunate" electrons in the avalanche, which do not collide with the phonons until an energy equal to the ionization potential I_i is accumulated [66, 67]. It is assumed here that the probability of the electron not colliding with the phonons is $\sim\exp(-I_i/eEl)$, where l is the electron mean free path, and that the collision frequency is independent of the field. This model is too simplified, although it does explain to some degree their experimental data on the depth and surface damage of various dielectrics. It should be noted, however, that surface damage is determined by the local electric field on the surface [68] and that various defects on the surface lead to a local increase of the electric field, thereby lowering the damage threshold [69]. The ex-

*We note that breakdown induced in alkali-halide crystals by CO_2 laser radiation was first investigated in [59].

periments reported in [65] were made on an ordinary surface whose defect structure was not investigated at all and was not taken into account in the calculation of the values of the effective field (defined by the authors as the square root of the intensity of the incident radiation), so that the results of their experiments cannot be regarded as proof of the dominant role of the electron avalanche in the damage mechanism. In Giuliano's opinion [22] the observed changes of the damage threshold from point to point are the consequence of differences in the optical endurance rather than a manifestation of the statistical character of the damage process.

A recent paper [70] reports investigation of damage of NaCl and KCl crystals at a wavelength of 10.6 μm. It is interesting that the KCl damage threshold observed in that study turned out to be higher than that of NaCl, and not conversely as reported in the earlier paper [57]. This casts doubt on the statement of the authors of [57, 68] that the correlation of the damage thresholds in the sequence of alkali-halide crystals is one of the principal arguments in favor of the electron avalanche mechanism of laser damage to these crystals.

Thus, the conclusion that the electron avalanche mechanism plays the decisive role in the damage of alkali-halide crystals [71], arrived at in [48, 49, 57] on the basis of, first, the correlation of the threshold fields of laser damage and of the breakdown in constant fields, second, the independence of the damage threshold of the electromagnetic radiation frequency, and, third, the statistical character of the damage process, cannot be regarded as proved. The question of the decisive damage mechanism of real transparent dielectrics therefore remains open; further research is called for.

2. LIGHT SCATTERING AT LOW INTENSITIES AND CRITERION OF OPTICAL PURITY OF TRANSPARENT MATERIALS

The foregoing analysis of the studies of laser damage to transparent dielectrics has shown that in most cases the damage is due to large (>0.1 μm) foreign inclusions. By improving the technology of the synthesis of the materials it is possible to get rid of their harmful influence on the optical endurance [56]. Recent communications report investigations of laser-damage mechanisms in sufficiently pure (containing no large inclusions) optical materials. In [48, 49, 57], which were discussed in detail above, the authors invesetigated damage to alkali-halide crystals in the absence of an influence of large absorbing defects and explained this damage on the basis of the mechanism of the electron avalanche that develops in a pure dielectric. However, the conclusion that the electron avalanche plays the dominant role cannot be regarded as sufficiently well founded, since no account was taken in these studies of the influence of minute (<0.1 μm) inclusions contained in the investigated crystals on the damage.

The purpose of our investigations was to ascertain the role of minute absorbing defects in the mechanism of laser damage to transparent materials. This made it necessary to estimate quantitatively the optical quality of the dielectrics from the point of view of their containing defects of this type.

Existing methods of estimating the optical quality do not make it possible to determine in a sufficiently simple manner the presence of extraneous inclusions and structure defects (dislocations, vacancy clusters, etc.). Observations of inclusions of size $\gtrsim 1$ μm, which can be seen with the aid of a microscope, are insufficient. An electron microscope cannot be used to investigate depth defects. X-ray structure analysis, while offering a number of advantages, is quite complicated. The method of decorating structure defects introduces considerable disturbances in the investigated object [72].

In contrast to the indicated method, the use of scattering light to investigate various types of defects in real transparent dielectrics makes it possible to determine relatively simply their optical quality. The most instructive method for estimates of the optical purity of a dielectric is, in our opinion, a comparison of the intensities of the scattering due to the defect structure of the substance (Rayleigh scattering) I_R and Mandel'shtam–Brillouin scattering (MBS) I_{MBS} in the same substance [40] or in some standard substance, inasmuch as such a comparison makes it possible to determine those scattering properties of the medium which are due to the presence of different defects (the number of such defects at a given dimension), without resorting to measurements of the absolute values of the scattering intensity, which are difficult to perform.*

We note that in real optical materials, even in very pure ones (containing no large defects and inclusions), the intensity of the Rayleigh scattering greatly exceeds as a rule the intensity of the MBS, thus attesting to the presence of a large number of defects and inclusions of small size. As to large inclusions, they can be easily observed by simpler methods and their role in laser damage has been well investigated [39]. We are interested in small absorbing defects, since it is precisely they which can determine the optical endurance limit of a real transparent dielectric. By minute absorbing defects and inclusions we mean here and elsewhere regions of small size with a local light-absorption coefficient greatly exceeding the absorption coefficient of the surrounding matrix. Thus, a detailed investigation of light scattering yields a quantitative estimate of the optical purity of real transparent dielectrics containing minute inclusions and defects.

Rayleigh and Mandel'shtam–Brillouin Scattering of Light in Transparent Media

The propagation of electromagnetic waves in real media is accompanied by their scattering and absorption. The cause of light scattering is optical inhomogeneity. This basic premise was established by Mandel'shtam back in 1907 [74]. The physical causes of optical inhomogeneities are numerous and varied. A medium can become optically inhomogeneous because of randomly distributed extraneous inclusions, structure defects (dislocations, vacancy clusters), and other types of microinhomogeneities.† Light scattering can be observed also in media that are completely free of any foreign inclusions, owing to the statistical character of the thermal motion of the particles of the medium, which causes fluctuations of the optical dielectric constant. Microinclusions or a foreign phase in transparent dielectrics can not only scatter the incident light, but also absorb it, and in the case of small particles the loss of light by absorption usually greatly exceeds the loss by scattering.

The intensity I_S of the scattered light depends not only on the physical characteristics of the scattering medium but also on the external conditions of the experiment: on the intensity of the incident light I_0, on the scattering volume V, and on the distance from the scattering volume to the point of observation L. To describe the scattering properties of a medium it is therefore customary to consider in place of I_S the scattering coefficient R [75], which does not depend on the experimental conditions: $R = \frac{I_s}{I_0}\frac{L^2}{V}$ (cm^{-1}), or the extinction coefficient h, which is connected with the scattering coefficient R_{90} of linearly polarized light observed at an angle 90° to the direction of propagation of the incident light: $h = \frac{8}{3}\pi R_{90}$.

For the scattering of linearly polarized light with wavelength λ, by a medium containing N scattering particles of radius $a \ll \lambda$ per unit volume, with a relative refractive index $m = n - i\chi$, the scattering coefficient R_{90} and the extinction coefficient h_R are calculated on the basis of the well-known Rayleigh formula [76]

$$R_{90} = \frac{\lambda^2}{4\pi^2}\left|\frac{m^2-1}{m^2+2}\right|^2 N\rho^6; \quad h_R = \frac{2\lambda^2}{3\pi}\left|\frac{m^2-1}{m^2+2}\right|^2 N\rho^6, \tag{1}$$

where $\rho \equiv 2\pi a/\lambda \ll 1$.

The scattering and extinction coefficients are similarly calculated also in the case of metallic particles for which $|m| \gg 1$ [75]:

$$R_{90} = \frac{5\lambda^2}{16\pi^2} N\rho^6; \quad h_R = \frac{5\lambda^2}{6\pi} N\rho^6. \tag{2}$$

*A similar comparison of the intensities of Rayleigh scattering and MBS, but in connection with calculation of the losses in materials used in fiber optics, was carried out in [73].
†We note that individual impurity ions, which are uniformly distributed in the volume of the medium, do not cause scattering of light in the medium.

To describe the total loss of light in the medium, both by scattering and by absorption by the scattering centers, one introduces the attentuation coefficient of the medium k_{att}, which, assuming not too small an imaginary part of the refractive index χ ($\chi > \rho^3$), is calculated from the formula [74]

$$k_{att} = \frac{6\lambda^2}{\pi} \frac{n\chi}{|m^2+2|^2} N\rho^3. \tag{3}$$

For small ρ, (1)-(3) describe the phenomena of scattering and absorption of light well, since the correction terms for them are of the order of ρ^2. Note here two important circumstances: 1) the attentuation coefficient of a small particle is proportional to its volume; this means that when light passes through a medium containing small particles the attenuation of the beam is proportional to the amount of particulate matter along the path of the light; 2) for small particles, the light lost to absorption at $\chi > \rho^3$ greatly exceeds the loss to scattering. The conclusions that follow from these facts will be considered below, in the discussion of the experimentally measured absorption and extinction coefficients in transparent dielectrics.

Scattering and extinction coefficients are introduced also for the description of the scattering of light by statistical inhomogeneities of the medium, which are connected with fluctuations of the dielectric constants [76]. We shall not dwell in detail on the concrete calculations of these coefficients in various transparent media. We present below only their experimentally measured values at the wavelength 6328 Å.

We estimate now the concentration of the scattering metallic particles with various dimensions in crystalline quartz. Comparing the extinction coefficients for the Rayleigh (R) and Mandel'shtam–Brillouin scattering (MBS) [76], we obtain

$$N\rho^6 = 5\cdot 10^2\, \eta, \tag{4}$$

where $\eta \equiv I_R/I_{MBS}$ is a parameter that characterizes the optical purity of the material. From (4) we can determine the particle concentration by specifying a definite dimension. We are interested in small particles with dimensions 100-1000 Å, which are usually present in pure materials. For example, for particles with dimension 300 Å we obtain at $\eta = 1$ from (4) the value $N \sim 10^7$ cm^{-3}. In real media, usually the parameter $\eta \gg 1$ (in particular, in the specially selected glasses investigated by us, $\eta \gtrsim 10^2$) and consequently the concentration of the scattering centers is much higher. It must be borne in mind here that Rayleigh scattering can be due to nonmetallic particles, and this leads to even higher concentrations of the scattering inhomogeneities at the same scattering intensity. Thus, in real media the concentration of the inclusions and defects that produce Rayleigh scattering turns out to be so high that even in the case of sharp focusing of the radiation in a focal volume of dimension 10^{-8}-10^{-7} cm^{-3} there are many such defects which can initiate the damage. It is apparently this circumstance which causes the absence of substantial variations of the damage thresholds from point to point.

A similar comparison of the intensities of the MB and Rayleigh scattering can be made in estimates of the optical quality of the surfaces of transparent dielectrics (the methods available to date do not make it possible to establish simply enough the presence of various types of defects in the surface layer of the sample). As indicated above, it is precisely the optical endurance of the surfaces of transparent materials which frequently limits the intensity of the transmitted light. This is caused by the presence of a surface layer with more defects, which turns out to be more highly scattering in comparison with the interior of the material if ordinary finishing methods are used. Elimination of the upper defective surface layer by ion polishing [22] or thermal evaporation [23] improves considerably the optical quality of the surface. This raises substantially the laser-damage threshold. We have investigated the so-called ideal surfaces of ruby, which contain no large defects [23]; the intensity of the Rayleigh scattering from these surfaces was comparable with the intensity of the scattering in the interior of the sample, and the threshold was much higher than for ordinary ruby surfaces treated by diamond polishing, which had a much higher scattering ability than the interior. Thus, our proposed criterion for optical purity can be used relatively simply and effectively in estimates of the optical quality of both the interior and the surface of a transparent material.

Experimental Investigation of the Optical Quality of Crystals and Glasses

Numerous investigations of the defect structure of glasses [77], ruby [78-80], and other transparent dielectrics [81] were made by the method of light scattering in them and by ultramicroscopy. It was found that the scattering centers influence both the threshold of laser damage of transparent materials [82] and the properties of the laser emission in the case when the scattering centers are present in active laser elements [83]. Thus, the optical quality of the employed transparent materials exerts a substantial influence on the course of various types of physical phenomena.

Different quantitative characteristics were proposed for the description of the optical quality of materials. The characteristic chosen in [77] was the coefficient of scattering at 90°, which made it possible to carry out relative measurements of the optical quality of different glasses, but did not make it possible to assess the purity of each material separately. In addition, it was found in that study that the light loss due to scattering is smaller by one-and-a-half orders of magnitude than the loss due to absorption. We note that a similar result was obtained also for light loss in ruby [79].

The method of ultramicroscopy using an He—Ne laser also effectively reveals microdefects in transparent dielectrics [81]. In this case the size of the particles is evaluated from the time of exposure on photographic film, and since the refractive index of the particles is not known, the scattering by the defects of the medium is measured in equivalent latex diameters (e.l.d.), i.e., the intensity of scattering by the defect is compared with that of scattering by latex spheres in water. The relative refractive index in this case is $m = 1.2$. It was established in [81] that an exposure time of 1 h makes it possible to detect scattering particles of size 300 Å e.l.d. The use of this procedure made it possible to find in ruby a strongly scattering region with dimensions ∿1000 Å e.l.d., decorated block boundaries with ∿700 Å e.l.d., and randomly disposed scattering centers with ∿400 Å e.l.d., [78]. In addition, there are also scattering defects of size <300 Å e.l.d., observable with the aid of photomultipliers and producing the greater part of the scattered light. Thus, ultramicroscopy makes it possible to observe defects in a medium which are much smaller than the wavelength of the incident light. However, even this mehod only makes it possible once again to assess the relative optical purity of transparent dielectrics measured in terms of equivalent latex diameters.

The most instructive method of estimating the optical quality of a dielectric is, in our opinion, a comparison of the intensities of the scattering due to defects in the medium with Mandel'shtam—Brillouin scattering in the same material. The MBS is due to the statistical character of the thermal motion of the particles of the medium, and is therefore completely determined by the state of the medium (its temperature, compressibility, and other parameters), is independent of the choice of the concrete sample of the given material, other external conditions being equal, and is a natural measure for the estimate of the optical quality of a dielectric. The optical purity of the various transparent materials, which served as objects for laser damage [84], was investigated on the basis of this proposed criterion. The scattering intensities of the MBS components were measured in all the materials relative to the scattering intensity of the longitudinal component in crystalline quartz 1,* for which the scattering coefficient at the wavelength $\lambda = 6328$ Å amounts to $R^{0}_{MB} = 0.5 \cdot 10^{-7}$ cm^{-1} [76].

The characteristics of the scattering of light and of the damage thresholds in crystals and glasses at pulse durations $\tau = 1.4$-7 nsec (for pure sapphire at $\tau = 2.5$-7 nsec) are listed in Table 1.

Measurement of the intensity of MBS in sapphire, which is weaker by one order of magnitude than MBS in quartz, was made difficult by the red luminescence of the impurities; the ratio η was therefore calculated in comparison with the intensity of the MBS in crystalline quartz. For an ideal crystal $\eta \simeq 0$, so that it can be assumed that in real crystals the observed Rayleigh scattering is due to scattering by defects. In glasses, the Rayleigh com-

*The experiments on the Rayleigh and Mandel'shtam—Brillouin scattering were performed by A. I. Ritus using a special setup with a scanning Fabry—Perot interferometer and photoelectric registration of the signal with the aid of a spectrum analyzer [85].

TABLE 1

Material	R_{MB}/R^0_{MB}	$\eta=I_R/I_{MB}$	Laser-damage threshold P_d, kW
Quartz			
crystal 1	1	0,12	—
crystal 2	1	3,3	110
Sapphire	—	1,2	130
Sapphire doped with Ni	—	17,0	12*
Glass			
K-8	0,5	100	100
BK-104	0,7	170	60
laser	0,7	90	60
TF-8	2,1	290	10

*The damage is due to absorbing inclusions with dimensions less than 0.1 μm.

ponent is due both to the inhomogeneity of the glass structure and to various kinds of inclusions. Although at the present time the question of the relative contribution of each of the indicated scattering sources remains open, nevertheless the parameter η in glasses can characterize the purity of the material. The results presented show that even sufficiently pure samples still contain a considerable number of defects. Such a high concentration of scattering defects gives one grounds for assuming that the light loss in an optical material (which usually amounts to $\sim 10^{-3}$ cm^{-1}, substantially exceeding the loss due to scattering, see Table 1) is due to a considerable degree to the absorption of the light by these defects. Estimates made with the aid of (1) and (3) show that in this case their light absorption coefficient should be quite large ($\sim 10^4$ cm^{-1}).

Thus, the question of the criterion of optical purity of transparent dielectrics, in connection with the problem of their laser damage, has up to now not been investigated or discussed in the literature. The existing methods of estimating the optical quality of the investigated materials are not sufficiently simple and effective. The proposed criterion of optical purity of transparent media that contain no large defects is based on a comparison of the intensity of the scattering due to the defect structure of the material with the Mandel'shtam—Brillouin scattering. This criterion can be used to estimate the optical quality of solid, liquid, and gaseous media. The experimentally measured intensities of the scattering by defects and of the MBS were used to estimate the purity of various crystals and glasses. The proposed purity criterion was used as the basis for the calculation of the concentration of minute (<0.1 μm) absorbing defects; this concentration turned out to be so high that even in the case of sharp focusing of the incident radiation into a focal volume measuring 10^{-8}–10^{-7} cm^3 their number is appreciable. Therefore consideration of the mechanisms of damage of real transparent dielectrics, without allowance for the inclusions and defects contained in them (as was done by a number of workers [48, 49]) is not correct enough, since these inclusions can initiate laser damage.

3. THEORY OF NONLINEAR THERMAL DAMAGE MECHANISM IN MEDIA WITH ABSORBING INCLUSIONS

A number of recent papers are devoted to the investigation of the mechanism of damage to transparent dielectrics containing absorbing inclusions [39, 86, 87]. This question was investigated in particular detail for the case of laser glasses with metallic and dielectric inclusions [39, 87]. It was experimentally established that the presence of platinum inclusions in the glass strongly influences the damage threshold. A similar effect was also observed for ruby laser crystals containing iron-group impurities (Ni, Co, Fe, Ti) [40].

The damage produced by absorbing inclusions and discussed in [39, 87] is due to the strong heating of the inclusions by the laser radiation. This heating can be accompanied by phase transitions within the inclusions (melting or evaporation) and by the onset of thermoelastic stresses in the surrounding matrix. In the cited papers there was developed a linear theory of thermoelectric damage, without allowance for the dependences of the parameters of the medium on the temperature, and the damage thresholds were estimated from the static values of the critical stresses. It is obvious that such an approach can lead to considerable errors for the following reasons. First, in this approach the values of the critical temperatures and stresses are not determined, in view of the dynamic character of the damage process (especially in the case of short laser pulses). Second, at such high temperatures ($\sim 10^4$ °K and above [39]), which develop in the course of the damage, an important role is played by the dependence of the thermal, elastic, and optical parameters of the medium on the temperature.

The possible role of the temperature dependences of the parameters of the medium in the damage process was indicated in [12, 87]. An attempt to take the temperature dependence of the absorption coefficient of the inclusions into account was made in [88].

We consider a more consistent theory of the thermal mechanism of the damage to transparent dielectrics, with account taken of the indicated dependences, both for the absorbing inclusions and for the surrounding matrix. In this approach, knowledge of the concrete values of the critical temperatures and stresses that lead to the damage is of no importance for the determination of the damage threshold. This permits a more realistic estimate of the thresholds of the depth damage of transparent materials containing absorbing inclusions.

Damage by Rectangular Laser Pulses

For simplicity we consider an absorbing spherical particle of radius a situated in a transparent dielectric. The process of heating such a particle and its surrounding medium by laser radiation is described by the heat-conduction equation*

$$\frac{\partial}{\partial t}(c\rho T) = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 k \frac{\partial T}{\partial r}\right) + Q(I, T), \tag{1}$$

where c, ρ, and k are the specific heat, density, and thermal conductivity coefficient, which are different for the inclusion and for the surrounding matrix, and which depend on the temperature; Q(I, T) is the strength of the thermal sources in the particle and depends on the intensity of the laser radiation and on the temperature T.

It must be borne in mind, however, that the equation of heat conduction in solids contains a term that depends on the elastic strains of the medium, and this term is usually neglected because of the small values of the strains [89]. When the thermal damage mechanism is considered, and the strains reach considerable sizes, allowance for them may turn out to be quite important and call for simultaneous solution of the heat-conduction and elasticity problems, thus raising considerable difficulties. We shall neglect the influence of the elastic stresses on the temperature distribution in the medium, and take into account the temperature dependence of the parameters of the medium.

For the temperature dependence of Q on the mechanism of absorption of the optical radiation, one can use various laws, e.g., the well-known approximation [90]

$$Q(I, T) = Q(I)\exp\left(\xi\frac{T - T_0}{T_0}\right), \tag{2}$$

where T_0 is the initial temperature of the sample; ξ is the parameter of the temperature nonlinearity of the absorption coefficient of the inclusion material. Such a dependence can take place, generally speaking, in a limited range of temperature variation.

The dependence of the coefficient of thermal conductivity on the temperature is different for different materials. We represent it in the form

$$k(T) = d/T, \tag{3}$$

where d is a constant of the material. Such a dependence is usually realized for dielectrics when T exceeds the Debye temperature θ and for a number of metals (e.g., nickel at T > 300 °K) [91].

The solution of Eq. (1) for $r < a$ and for times $t > \tau_X = T_0 c_1 \rho_1 a^2/\gamma_1$ (the index 1 pertains to the inclusion material, and 2 pertains to the surrounding matrix) will be sought in the form

$$T(r, t) = T_0 \exp[U(t)(\beta - r^2/3a^2)], \tag{4}$$

*Such an analysis is valid for particles that are not too small, when the size effects play no important role in the phonon spectrum.

where $\beta = {}^1/_3(1 + 2d_1/d_2)$; U(t) is a certain function that changes negligibly over a time τ_x. This approximation is valid under the condition $\tau_x \frac{dT}{dt} \ll T - T_0$. Substituting (4) in Eq. (1), we obtain for the temperature at the center of the inclusion at the instant of time t the implicit function

$$t = \int_{x_1}^{x_2} \frac{T_0 \beta e^{\beta x} C_1 (T_0 e^{\beta x})\, \rho_1 (T_0 e^{\beta x})\, dx}{Q(I, T_0 e^{\beta x}) - 2d_1 x/a^2}, \tag{5}$$

where

$$x_1 = \frac{a^2}{2d_1} Q(I, T_0); \qquad x_2 = \frac{1}{\beta} \ln T/T_0.$$

Choosing relation (2) for Q(I, T), assuming C_1 and ρ_1 to be independent of temperature, and taking into account the rapid convergence of integral (5) we can set its upper limit equal to infinity. Then

$$t_{\text{thr}} = \tau_x \int_{x_1}^{\infty} \frac{\beta e^{\beta x} dx}{d_1^{-1} Q_{\text{thr}}(I)\, a^2 \exp[\xi (e^{\beta x} - 1)] - 2x}. \tag{6}$$

In this case the quantity t_{thr} has the meaning of the time after which the temperature at the center of the inclusion begins to increase very rapidly. The maximum attainable temperature in the particle can be sufficiently high and is determined either by the saturation of the temperature nonlinearity of the absorption, or by the energy lost to the thermal radiation. This temperature determines the maximum stresses in the surrounding matrix, and consequently also the character, magnitude, and dynamics of the damage produced in the matrix. However, even without knowing the maximum temperatures and stresses it is possible to determine the damage threshold condition. Indeed, the rapid avalanche-like growth of the inclusion temperature, which precedes the saturation effect and follows from Eq. (6), means that the time t_{thr} can be regarded as the start of the damage, and the quantity Q_{thr} can be regarded as the threshold damage power. We assume in this case that on the linear section of the growth of the inclusion temperature (at $t < t_{\text{thr}}$) no damage takes place. This is attested to by the fact that the damage produced in transparent dielectrics by laser radiation always has a clearly pronounced threshold and is accompanied by high-temperature radiation (spark). The presence of such a threshold emission fits well within the framework of the considered model of the nonlinear thermal damage mechanism, and cannot be explained by the existing linear theories [39, 86, 87]. Thus, the damage process in the considered model has the character of a thermal explosion.

Formula (6) determines the dependence of the threshold damage power on the duration of the laser pulses. In particular, for rectangular pulses of duration τ this dependence, calculated by computer for several values of the parameters ξ and β, is shown in Fig. 1.

In the case when $k(T) = d_1/T$ and the temperature nonlinearity of the absorption is weak, we can confine ourselves in (2) to the first two terms of the series expansion, and we obtain for the temperature at the center of the inclusion

$$\frac{T(t) - T_0}{T_0} \simeq \frac{Q(I)\, a^2}{d_1 \nu} \left[1 - (1 - \nu) \exp\left(-\frac{t\nu}{\tau_x}\right)\right], \tag{7}$$

where

$$\nu = \frac{Qa^2}{d_1}(\xi - 1) - \frac{2}{\beta} \ln \xi.$$

It is seen from this formula that at

$$\frac{Qa^2}{d_1} |\xi - 1| > \frac{2}{\beta} |\ln \xi|$$

the temperature of the inclusion begins to increase exponentially with time, corresponding in fact to thermal explosion. It follows therefore that at laser pulse duration $\tau \gg \tau_x$ we have

$$Q_{\text{thr}}(I) = \frac{2d_1}{a^2 \beta} \frac{\ln \xi}{\xi - 1}. \tag{8}$$

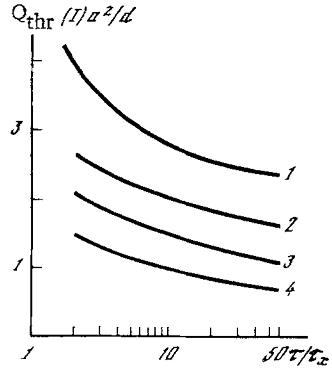

Fig. 1. Dependence of the threshold damage power on the laser pulse duration: 1) $\beta = 1$, $\zeta = 0.1$; 2) $\beta = 2.5$, $\zeta = 0.1$; 3) $\beta = 1$, $\zeta = 0.2$; 4) $\beta = 2.5$, $\zeta = 0.2$.

Under the same conditions it is easy to obtain from (1) the threshold damage conditions in the linear case, when the parameters of the medium do not depend on the temperature, assuming that the damage sets in before a certain critical temperature T_{cr} is reached. In this case

$$Q_{thr}(I) = \frac{2d_1}{a^2\beta}\frac{T_{cr}}{T_0}. \tag{8a}$$

Comparison of (8) and (8a) shows that when account is taken of the temperature dependence of the parameters of the medium the damage threshold can be determined without knowing the value of T_{cr}, as in the linear theory. The damage threshold is determined not by the ratio T_{cr}/T_0 but by the form of the nonlinearity and by the value of the corresponding parameter, quantities that can be experimentally determined by investigating the temperature dependence of the coefficient of light absorption by the material of the inclusions if the latter are not too strongly heated. From a comparison of these formulas it follows that for not too small a nonlinearity parameter $\xi > 10^{-4}$, at one and the same dimension of the inclusion, the damage threshold is much smaller in the nonlinear case (the ratio of the thresholds in both cases being determined only by the form of the temperature dependence of the light absorption by the medium) or, equivalently, in the nonlinear case the same damage threshold is obtained for inclusions of much smaller size than in the linear case.

To determine the laser emission threshold intensities that lead to damage it is necessary to know the $Q_{thr}(I)$ dependence, which can be nonlinear. In the case of the usual linear (single-photon) absorption, for particles of small size ($a \ll \lambda$), using the known expression for the absorption for the cross section [92], we can express the threshold intensity I_{thr} in the form

$$I_{thr} = \frac{\lambda}{18\pi}\frac{(\varepsilon' + 2)^2 + \varepsilon''^2}{\varepsilon''} Q_{thr}, \tag{9}$$

where $\varepsilon = \varepsilon' + i\varepsilon''$ is the relative dielectric constant of the particle material at the wavelength λ. This formula does not take into account the magnetic part of the absorption, which can make an appreciable contribution in the case of metallic particles having high conductivity.

The foregoing analysis of the damage process pertains to the case of "long" laser pulses ($\tau > \tau_x$). For short pulses ($\tau \ll \tau_x$) in the case of uniformly absorbing particles we obtain from (1)

$$Q_{thr} = \frac{C_1\rho_1 T_0}{\xi\tau}\left\{1 - \exp\left[-\xi\left(\frac{T_{cr}}{T_0} - 1\right)\right]\right\}, \tag{10}$$

where T_{cr} is the critical temperature of the particle at which the damage sets in. Since $T_{cr} \gg T_0$, for not too small a nonlinearity parameter ξ and in the case of short pulses we can also estimate Q_{thr} without knowing T_{cr}.

We now estimate the threshold values Q_{thr} and I_{thr} for ruby crystals containing metallic nickel particles. Such particles (as well as particles of other metals) can be present in ruby laser crystals [40]. We take by way of example a typical particle dimension $2a = 3 \cdot 10^{-6}$ cm and a laser pulse $\tau = 3 \cdot 10^{-8}$ sec. In this case $\tau \gg \tau_\chi$ and to estimate Q_{thr} we must use Eqs. (6) and (8). We take for the parameter ξ the value $\xi \simeq 0.1$ (this is approximately the value measured by us for films of nickel and of the oxides NiO, CoO, and FeO in the temperature range 300-1500°K at a wavelength $\lambda = 0.63$ μm). Taking for ε' and ε'' the known values [92], we obtain from (6) and (9) $Q_{thr} \simeq 10^{14}$ W/cm³ and $I_{thr} \simeq 2 \cdot 10^9$ W/cm². Estimates from (8) and (9) for a weaker nonlinearity yield $Q_{thr} \simeq 3.5 \cdot 10^{14}$ W/cm³ and $I_{thr} \simeq 7 \cdot 10^9$ W/cm². The foregoing estimates of I_{thr} correspond approximately to the observed values of the damage threshold of sapphire samples containing nickel inclusions of the indicated size [40].

As estimate of the threshold damage power of glass containing platinum inclusions of dimension $2a = 10^{-5}$ cm, produced by a laser pulse of duration $\tau = 3 \cdot 10^{-8}$ sec under the assumption that $\xi = 0.1$, yields $I_{thr} = 8 \cdot 10^7$ W/cm². This value is smaller by one order of magnitude than that calculated in [39] on the basis of the linear theory.

Damage by Bell-Shaped Laser Pulses

We start with the heat-conduction equation for the entire medium, assuming that k and χ, the coefficients of thermal conductivity and thermal diffusivity, respectively, do not depend on the temperature T and are the same for the entire medium:

$$\frac{\partial T}{\partial t} = \chi \nabla^2 T + \frac{\chi}{k} Q \tag{11}$$

under the following initial and boundary conditions: $T = 0$ at $t = 0$; $T \to 0$ and $\nabla T \to 0$ as $r \to \infty$. In Eq. (11) the power Q of the absorbing sources will be assumed in the form

$$Q = Q_0(\mathbf{r})\,\theta(t)\,F(T), \tag{12}$$

where the function $Q_0(\mathbf{r})$ describes the coordinate dependence; $\theta(t)$ is the time-dependent form of the incident radiation pulse and satisfies the conditions

$$\theta(t) \leqslant 1,\ \theta(0) = 0,\ \theta'(0) = 0. \tag{13}$$

The function F(T) describes the temperature dependence of the light absorption by the medium.

We first consider the case when the light absorption by the medium is independent of temperature: $F(T) = 1$. In this case the solution of Eq. (11), with allowance for the conditions (12) and (13), is written in the form [41]

$$T(\boldsymbol{r}, t) = \frac{1}{\pi^{3/2}} \frac{\chi}{k} \int_0^t \int_V Q_0(\rho)\,\theta(\tau)\,\varepsilon(\boldsymbol{r} - \rho, t - \tau)\,dV\,d\tau, \tag{14}$$

where $\varepsilon(\mathbf{r} - \rho,\ t - \tau)$ is the fundamental solution of the heat-conduction equation: $\varepsilon(\boldsymbol{r} - \rho, t - \tau) = \frac{1}{[4\chi(t-\tau)]^{3/2}} \exp\left[-\frac{|r-\rho|^2}{4\chi(t-\tau)}\right]$. From (14), using the rule for differentiating a convolution [94] and taking conditions (13) into account we can calculate the values of the first and second derivatives of the temperature with respect to time. As a result we have

$$\frac{\partial T}{\partial t}(\boldsymbol{r}, t) = \frac{1}{\pi^{3/2}} \frac{\chi}{k} \int_0^t \int_V Q_0(\rho)\,\theta'(\tau)\,\varepsilon(\boldsymbol{r} - \rho, t - \tau)\,dV\,d\tau, \tag{15}$$

$$\frac{\partial^2 T}{\partial t^2}(\boldsymbol{r}, t) = \frac{1}{\pi^{3/2}} \frac{\chi}{k} \int_0^t \int_V Q_0(\rho)\,\theta''(\tau)\,\varepsilon(\boldsymbol{r} - \rho, t - \tau)\,dV\,d\tau. \tag{16}$$

From (14)-(16) it is seen that the time dependence of T is "stretched-out" relative to the incident-radiation pulse, i.e., the maximum of T is reached after the maximum of the radiation pulse, and the growth rate of T, determined by the second derivative of T with respect to time from (16), slows down later than the rate of change of the incident pulse, and the value of this "stretch-out" depends on the ratio of the duration of the laser pulse to the characteristic heat propagation time: $\tau_\chi = a^2/4\chi$. The dependence of T on the time for different pulse durations τ is shown schematically in Fig. 2. It must be emphasized that in the case when the power of the absorbing sources does not depend on the temperature a maximum

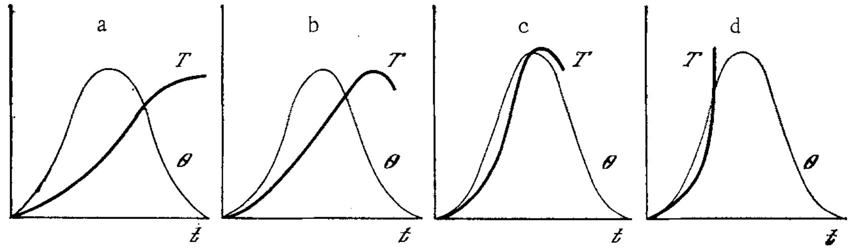

Fig. 2. Heating of an absorbing defect at different durations of bell-shaped laser pulses: a-c) in the case when the absorption of light by the medium does not depend on the temperature, and d) in the case when account is taken of the temperature dependence of the parameters of the medium. a) $\tau < \tau_X$; b) $\tau = \tau_X$; c) $\tau > \tau_X$; d) $\tau > \tau_X$.

of the temperature is always reached. This means that within the framework of the linear theory of the thermal damage mechanism we must know the critical temperatures and stresses in order to determine the threshold. In addition, the approach to this not-fully-determined critical temperature with increasing laser power has a smooth character and cannot explain the presence of a threshold for the high-temperature emission that accompanies the damage. Let us analyze the case when the coefficient of light absorption by the medium depends on the temperature. We write the solution of Eq. (11), with allowance for the conditions (12) and (13), in the form

$$T(\boldsymbol{r}, t) = \frac{1}{\pi^{3/2}} \frac{\chi}{k} \int_0^t \int_V Q_0(\rho)\, \theta(\tau)\, F\,[T(\rho, \tau)]\, \varepsilon(\boldsymbol{r} - \rho, t - \tau)\, dV\, d\tau. \tag{17}$$

In analogy with (15) and (16) we obtain the derivatives of T with respect to time:

$$\frac{\partial T}{\partial t}(\boldsymbol{r}, t) = \frac{1}{\pi^{3/2}} \frac{\chi}{k} \int_0^t \int_V Q_0 \theta F \left[\frac{\theta'}{\theta} + \frac{F'}{F}\frac{\partial T}{\partial \tau}\right] \varepsilon(\boldsymbol{r} - \boldsymbol{\rho}, t - \tau)\, dV\, d\tau, \tag{18}$$

$$\frac{\partial^2 T}{\partial t^2}(\boldsymbol{r}, t) = \frac{1}{\pi^{3/2}} \frac{\chi}{k} \int_0^t \int_V Q_0 \theta F \left[\frac{\theta''}{\theta} + 2\frac{\theta'}{\theta}\frac{F'}{F}\frac{\partial T}{\partial \tau} + \frac{F''}{F}\left(\frac{\partial T}{\partial \tau}\right)^2 + \frac{F'}{F}\frac{\partial^2 T}{\partial \tau^2}\right] \varepsilon\, dV\, d\tau, \tag{19}$$

where F' and F" are the derivatives of F with respect to temperature, which depend on T and are determined by the form of the nonlinearity of the absorption. We assume a nonsaturating nonlinearity, i.e., such that $F' > 0$ and $F'' > 0$. As shown above, within the framework of the nonlinear theory of the thermal damage mechanism, the criterion of the threshold is a rapid growth of the temperature with time in the course of heating by the laser pulse. On the basis of this criterion, above we determined the threshold of the laser damage for the case of rectangular radiation pulses. We obtain analogously the threshold conditions for the case of bell-shaped laser pulses. It is obvious that the condition of thermal explosion is that the second derivative of the temperature with respect to time be positive ($\partial^2 T/\partial t^2 > 0$). Let us determine at what parameters of the problem this condition can be reached. The values of the functions Q_0, θ, F, ε in the entire integration interval are larger than zero, and whether $\partial^2 T/\partial t^2$ in (19) is positive or negative depends on the sign of the expression in the square brackets. It is clear that the fourth term in this expression is greater than zero. We introduce the notation

$$\frac{F''}{F} = p; \quad \frac{\theta'}{\theta}\frac{F'}{F} = q; \quad \frac{\theta''}{\theta} = r; \quad \frac{\partial T}{\partial \tau} = x.$$

The condition $\partial^2 T/\partial t^2 > 0$ is then reached at

$$X = px^2 + 2qx + r \geqslant 0. \tag{20}$$

We consider separately several time intervals:

1) $0 < \tau < t''$, where t" is determined by the condition $\theta''(t'') = 0$. Here $p > 0$, $q > 0$, $r > 0$ and consequently $X > 0$.

2) $t'' < \tau < t'$, where t' is determined from the condition $\theta'(t') = 0$. Here $p > 0$, $q > 0$, $r < 0$; therefore $X > 0$ at

$$\frac{\partial T}{\partial \tau} > \frac{\sqrt{q^2 - pr} - q}{p}. \tag{21}$$

Thus, the thermal explosion condition ($\partial^2 T/\partial t^2 > 0$) is satisfied on the leading front of the laser pulse when the condition (21) is satisfied. This condition can be reached at a definite (threshold) laser radiation power. To make the results more illustrative, we consider the particular case of a laser pulse with a time waveform $\theta(t) = \left(\frac{t}{\tau}\right)^2 \exp\left[-\left(\frac{t}{\tau}\right)^2\right]$ and an absorption nonlinearity temperature in the form $F(T) = \exp\left[\xi \frac{T - T_0}{T_0}\right]$. In this case

$$p = \xi^2/T_0^2; \quad q \simeq \frac{2\xi}{T_0 \tau} \frac{1 - (t/\tau)^2}{t/\tau}; \quad r = \frac{2}{\tau^2} \frac{1 - 5(t/\tau)^2 + 2(t/\tau)^4}{(t/\tau)^2};$$
$$t' = \tau; \; t'' \cong 0.47\tau$$

and condition (21) takes the form

$$\frac{\partial T}{\partial t}(t) > \frac{T_0}{\xi \tau} \frac{\sqrt{2[1 + (t/\tau)^2]} - 2[1 - (t/\tau)^2]}{t/\tau}. \tag{22}$$

It is satisfied if $\frac{\partial T}{\partial t}(t'') > \frac{2T_0}{\xi\tau}$.

An estimate of the threshold power of the absorbing sources, for the case of an inclusion of radius a and a pulse duration $\tau \gg \tau_\chi$, yields $Q_0^{thr} \simeq 4kT_0/\xi a^2$. This result is somewhat too high, and it can be regarded only as an order-of-magnitude estimate. The threshold dependence of the laser, just as in the case of rectangular pulses, is determined in accordance with (9).

In the case of short laser pulses $\tau \ll \tau_\chi$ we can neglect the heat dissipation in Eq. (11). For a laser pulse $\theta(t) = (t/\tau)^3 \exp[-(t/\tau)^2]$ we obtain therefore

$$T - T_0 = -\frac{T_0}{\xi} \ln\left[1 - \frac{\xi}{T_0} \frac{Q_0 \chi \tau}{2k} \theta_1(t)\right], \tag{23}$$

where $\theta_1(t) = 1 - \exp[-(t/\tau)^2][1 + (t/\tau)^2]$. It is seen from (23) that at $Q_0^{thr} = 2kT_0/(\xi\chi\tau\theta_1(\tau))$ the temperature increases without limit, a situation corresponding in fact to thermal explosion.

An analysis of the solutions of Eq. (11) for the case of short laser pulses of rectangular and bell shapes shows that the threshold radiation intensity I_d depends to a considerable degree on the concrete form of the temperature dependence of the parameters of the medium. However, in contrast to rectangular laser pulses, when $I_d \sim 1/\tau$ (i.e., the damage threshold is determined by the energy of the laser pulse), in the case of bell-shaped pulses I_d is determined not only by the pulse duration but also by its waveform. This makes the dependence of I_d on τ more complicated.

The foregoing analysis of the case of bell-shaped laser pulses makes it easier to explain the presence of a threshold high-temperature emission accompanying the damage process, as well as the fact that the damage almost always takes place on the leading front of the laser pulse. In addition, it follows from the presented results that the damage threshold is determined not only by the peak power of the incident radiation, but also by the slope of the leading front of the radiation pulse.

The theory developed here for thermal damage of transparent dielectrics containing absorbing inclusions shows that the damage has the character of thermal explosion. This enables us to estimate the threshold damage conditions for different classes of transparent dielectrics (glass, crystals) without analyzing the dynamics of the development of the thermoelastic stresses. The theory shows that the damage threshold depends substantially on the dimension of the inclusions, on the thermal and optical constants, and on the temperature dependences. Although in the presented analysis we have confined ourselves to particular cases of temperature dependences of the thermal conductivity and of the absorption coefficient, it is obvious that a similar analysis can be carried out also for other types of nonlinearity.

The damage theory considered by us is quite general, since it is valid for any light-absorption mechanism that leads to a quasiequilibrium heating of the phonons (the condition for the concept of instantaneous temperature of the lattice). One of these mechanisms is multiphoton absorption by free carriers produced in avalanche ionization of the lattice or of the impurity centers.

Thus, the foregoing analysis of the damage mechanism can be used to analyze the experimental data on depth damage of transparent dielectrics in a wide range of laser pulse durations [95]. In particular, it can explain the large scatter of the published data (see, e.g., [21]) on the damage thresholds of glasses and crystals, which is due to the strong dependence of the threshold on the dimension of the absorbing inclusions and their nature.

4. EXPERIMENTAL SETUPS AND METHODS OF INVESTIGATING LASER DAMAGE

Laser damage to transparent dielectrics was investigated in many studies, a brief review of which is contained in Sec. 1. An analysis of these studies shows that the experimental results are determined to a considerable degree by the investigation procedures used and by the characteristics of the output radiation of the lasers employed. It was shown in a number of papers [51, 53, 54] that the damage process can depend substantially on the characteristics of the laser radiation, particularly on its mode composition and on the spatial distribution of the intensity in the beam cross section. It is well known, e.g., that in those cases when the damage goes through a self-focusing stage the character of the damage is different when single-mode and multimode lasers are used (in the case of multimode radiation, series of parallel damage tracks are observed, while a single track is observed in the case of single-mode lasers).

The investigation procedures determined also to a considerable degree the success in the study and understanding of the mechanisms and processes of damage of transparent dielectrics. Methods in which the onset of the damage is revealed by the appearance of a high-temperature spark at the breakdown instant, and by visible mechanical damage of the material after the passage of the radiation pulse, yield no information whatever concerning those processes of interaction of radiation with matter that lead to the damage. The most complete information concerning these interactions is provided by research methods that use dynamic effects that accompany the damage process. The latter include nonlinear scattering of light near the laser damage threshold (at intensities lower as well as higher than threshold), the glow accompanying the damage process, and the absorption of the energy of the passing radiation pulse. The question of the procedures used to investigate laser-induced damage processes will be examined below.

The use of single-mode lasers permits the simplest and most adequate study of the damage processes and the interpretation of the results. The possible use of single-mode radiation to identify the damage mechanism became obvious after it was demonstrated that the damage depends on the mode composition [53, 54]. Therefore in the investigation of the possible damage mechanisms in sufficiently pure materials we used laser radiation with spatially uniform intensity distribution. However, in the study of certain aspects of the damage problem (in particular, when one assesses the role of different impurities in the same material), multimode lasers can also be used.

Ruby Laser for the Investigation of Damage at Radiation Pulse Duration from 20 to 40 nsec

In the investigation of certain aspects of the damage problem, we used a ruby laser system (Fig. 3) consisting of a master generator operating in the Q-switched regime with saturating filter and a recording system.

The active element of the laser was a ruby crystal (l = 80 mm, d = 14 mm) pumped by four xenon flash lamps of type IFP-800. The exit mirror of the resonator was a stack of plane-parallel quartz plates, having a reflection coefficient of the order of 30% for light of wavelength λ = 6943 Å. The saturating filter was a solution of cryptocyanine in nitrobenzene. By varying the concentration of the solution and the length of the resonator it was possible to change the duration of the output pulses of the master generator in the 20-40-nsec range at half-height. The pulses were smooth and bell-shaped. The output power of the master generator was maintained constant, and its deviations from the mean value did not exceed ±10%. The output power and the duration and pulse waveform of the laser radia-

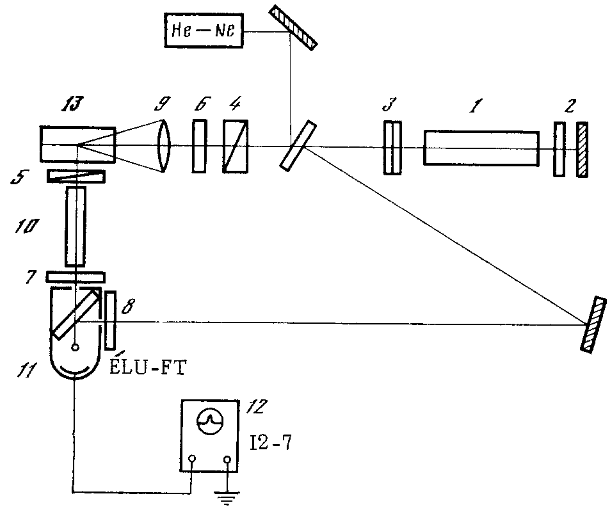

Fig. 3. Ruby laser system with radiation pulse duration 20-40 nsec: 1) ruby crystal; 2) nonlinear absorbing filter; 3) stack of plane-parallel quartz plates; 4) Glan prism; 5) polaroid; 6-8) sets of neutral light filters; 9) focusing lens; 10) light pipe; 11) electron beam amplifier; 12) oscilloscope; 13) investigated sample.

tion were monitored with an electron-beam multiplier ÉLU-FT with resolution not worse than $3\cdot10^{-9}$ sec, and with a high-speed I2-7 oscilloscope. To obtain a single-mode lasing regime, a diaphragm of diameter $\simeq 1$ mm was placed inside the resonator.

To investigate various processes that accompany laser damage to transparent materials, we used the following optical elements in the recording system. A Glan prism and a polaroid made it possible to measure the intensities of the transmitted light or of the scattering by the method of crossed and noncrossed polaroids. The intensity of the laser radiation was attenuated with calibrated neutral light filters. The radition was focused in the interior of the sample with aid of a lens of focal length F = 5 cm. The scattered light and the glow accompanying the damage were registered at an angle 90° to the direction of the incident radiation by means of an electron-beam multiplier ÉLU-FT and an I2-7 oscilloscope. The light filters KS-19 and SZS-22, 5 mm thick, made it possible to observe separately the scattered light and the luminescence. An optical delay line of $\simeq 50$ nsec made it possible to observe simultaneously on the oscilloscope screen the pulses of the incident and scattered radiation or of the luminosity (or of the transmitted light if the receiver was placed behind the sample), thus eliminating the influence of the instability of the laser output power. The probing He—Ne laser was used to indicate the possible presence in the sample of large defects and inclusions (thereby excluding also their influence on the damage) and the onset of mechanical damage in the material.

Single-Frequency Ruby Laser with Spatially Homogeneous Radiation Field and with Variable Pulse Duration in the Nanosecond Band

In the investigation of the possible damage mechanisms in sufficiently pure materials it is necessary to use a laser with a spatially homogeneous radiation field. This is usually done by producing single-mode radiation in the lowest mode TEM_{00} with the aid of standard mode selection within the resonator followed by amplification. However, it is difficult to obtain high-power single-mode radiation by this method in solid-stage lasers, owing to the optical inhomogeneity of the crystals used in the generators and amplifiers, which makes the operation of such laser systems unstable. The single-mode regime is stable in this case only at slight excess of the pump power over threshold. We describe below a new method proposed by us for obtaining high-power laser radiation with spatially homogeneous distribution of the intensity in the beam cross section; this method is not sensitive to the quality of the laser crystals and to the excess of pump power over threshold.

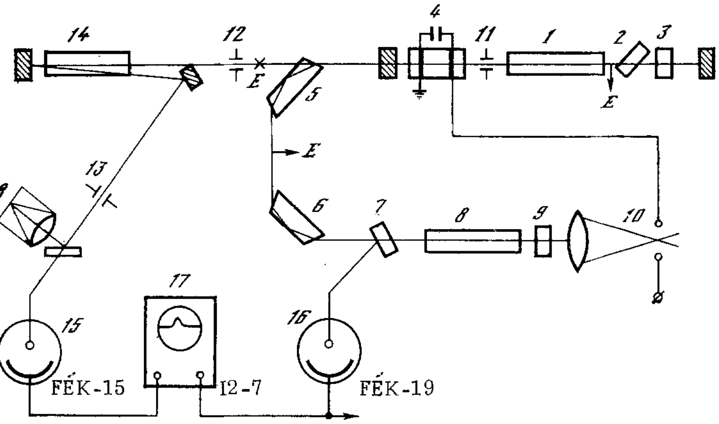

Fig. 4. Block diagram of single-frequency ruby laser with variable pulse duration: 1) ruby crystal; 2) nonlinear absorbing filter; 3) selector of longitudinal oscillation modes; 4) electrooptical shutter; 5, 6) Brewster prisms; 7) beam-splitting plate; 8) amplifying ruby crystal; 9) light filters; 10) high-pressure discharger; 11) diaphragm; 12, 13) diaphragms of distributed filters; 14) ruby crystal; 15, 16) photorecievers; 17) oscilloscope; 18) calorimeter.

The overwhelming number of studies were made on laser damage at radiation pulse durations from 20 to 30 nsec. In the damage problem, however, light pulses with variable duration in the nanosecond and subnanosecond bands from 0.1 to 10 nsec are interesting, inasmuch as the characteristic times of establishment of various nonlinear processes that play an important role in laser damage can lie in this interval. We therefore performed experiments in the pulse duration range 1-10 nsec.

To obtain short laser pulses in the nanosecond band one can use the method of introducing the radiation from a closed resonator (the Vuylsteke method [96]). However, the use of resonator length variation to vary the pulse duration in this scheme is not convenient in practice and, furthermore, when the resonator tuning is changed the output characteristics of the laser (the frequency spectrum, the spatial intensity distribution, and the radiation power) are altered.

A new high-power ruby laser with variable pulse duration in the nanosecond band was developed [97], using, in contrast to the Vuylsteke method, an exceedingly simple electric method of varying the radiation pulse duration without changing the laser configuration. The schematic diagram of such a laser is shown in Fig. 4. The laser system consists of two parts, the master generator in which the short light pulses are formed, and the power amplifier with distributed filters.

The master generator operates in the Q-switching regime with a saturable filter and with transmission coefficient 21%. The active element of the laser is a ruby crystal (l = 80 mm, d = 14 mm), pumped by four IFP-800 xenon flash lamps. The mirrors of the resonator have reflection coefficients of 97 and 90%. To select the longitudinal modes, a stack of plane-parallel quartz plates is used in the resonator. We note that effective longitudinal-mode selection is possible also without a stack, by precision adjustment of the crystal and the resonator mirrors. A diaphragm of $\simeq 3$ mm diameter is placed in the resonator to limit the size of the output beam and to improve the operation of the electrooptical shutter, by decreasing the influence of the optical inhomogeneity of the KDP crystal and of the inhomogeneity of the electric field. The master generator operates in a regime in which there are many transverse modes. The radiation pulse duration is $\simeq 50$ nsec.

The short radiation pulses are obtained with a single-block electrooptical Pockels shutter using a KDP crystal operating on the basis of the birefringence effect [98]. The electric supply circuit for the shutter consists of a high-voltage source (24 kV), cable lines, and a nitrogen-filled high-pressure discharger (9 atm) controlled by the optical radiation of the laser, which passes first through an amplifier (ruby crystal with dimensions l = 120 mm and d = 8 mm, pumped with the aid of two xenon flash lamps of type IFP-

2000). The position of the section clipped out of the master laser pulse is regulated by the cable delay lines and by the use of light filters to vary the intensity of the light entering the dicharger. A rectangular voltage pulse with amplitude equal to double the opening voltage is applied to the Pockels shutter. This produces on the leading front of the control-voltage pulse a radiation pulse whose duration and waveform are determined by the slope of the front. On the trailing edge of the control pulse there is also produced a radiation pulse whose duration and waveform are determined by the slope of the trailing edge, if the generation of the giant pulse inside the master laser still goes on at that instant. Consequently, to obtain single laser pulses it is necessary that the voltage pulse be longer than the lasing pulse of the master laser. We note that the production of paired light pulses with variable duration and with variable time delay between them can be of interest for certain applications. Variation of the laser pulse duration is effected in this system by blocking up the front of the voltage pulse with the aid of noninductive capacitors connected to the strip electrodes of the Pockels shutter. We emphasize that this electric method of varying the radiation pulse duration without changing the configuration of the laser is exceedingly simple compared with other methods (e.g., in Vuylsteke's scheme the pulse duration is varied by changing the resonator length, which is not convenient in practice).

We note that the same formation of laser pulses can be effected by clipping them from the output radiation pulse with the aid of a shutter located outside the resonator. However, the power of the clipped pulse is in this case much lower than in the case of extraction from the closed resonator.

The shaped radiation pulse is extracted from the closed resonator to the outside of the exit mirror at an angle φ of the optical axis of the master laser with the aid of an electrooptical shutter based on the birefringence effect. The angle is determined by the concrete construction of the shutter and in our case was $\simeq 2°$. The radiation extracted from the resonator is further amplified by a two-pass power amplifier 14 (ruby crystal with dimensions l = 240 mm and d = 12 mm, pumped by four IFP-5000 xenon flash lamps). To obtain spatially homogeneous output radiation with a diffraction divergence, an input diaphragm 12 and an output diaphragm 13 are used with respective diameters 0.8 and 1 mm, spaced $\simeq 3.5$ m apart. The distance between the diaphragms and the amplifying ruby crystal and also the dimensions of the diaphragms are determined from the following considerations: As indicated above, the master laser operates in a regime with many transverse modes, corresponding to radiation with an inhomogeneous phase front and with an inhomogeneous amplitude distribution. When the laser radiation is diffracted by the entrance diaphragm, the small-scale inhomogeneities of the field are effectively smoothed out. By placing the exit diaphragm in the wave zone of the entrance diaphragm (at a distance $\gg 1$ m), it is possible to make the characteristic dimension of the field inhomogeneity at the exit diaphragm much larger than the diaphragm diameter. In this case the distribution of the intensity in the cross section of the beam that passes through the exit diaphragm corresponds to the diffraction pattern of radiation with a plane phase front diffracted by a round hole. It must be borne in mind, however, that owing to the inhomogeneity of the ruby crystal used as a power amplifier, additional distortions are introduced into the phase front; therefore the exit diaphragm must be placed with these distortions taken into account. Thus, this system of diaphragms produces spatial filtering by causing high homogeneity and diffraction divergence of the exit radiation, which is equivalent to single-mode laser emission.

The following characteristics of the radiation of the laser system produced by us were investigated: the spatial distribution of the intensity, the time shape of the pulse, and the radiation power. The distribution of the intensity in the beam cross section in the far zone of the exit diaphragm was very uniform (Fig. 5), and its divergence amounted to $\simeq 10^{-3}$ rad, corresponding to the diffraction limit.

The waveform of the radiation pulse was registered with the aid of an FÉK-15 photomultiplier and I2-7 high-speed oscilloscope. The duration of the light pulse in our setup was varied between 1.4 and 7 nsec. Typical oscillograms of the laser pulses are shown in Fig. 6. As seen from Fig. 6, the pulses remain smooth when the duration is varied in a wide range. With increasing pulse duration $\tau \gtrsim 2L/c$ (L is the optical length of the resonator), their form becomes distorted. Distortions of this type were observed in our installation at $\tau \gtrsim 7$ nsec.

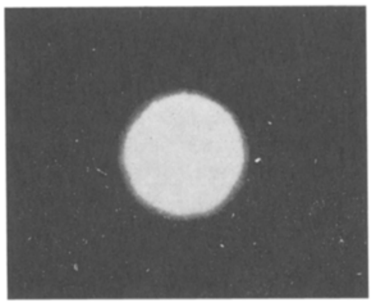

Fig. 5

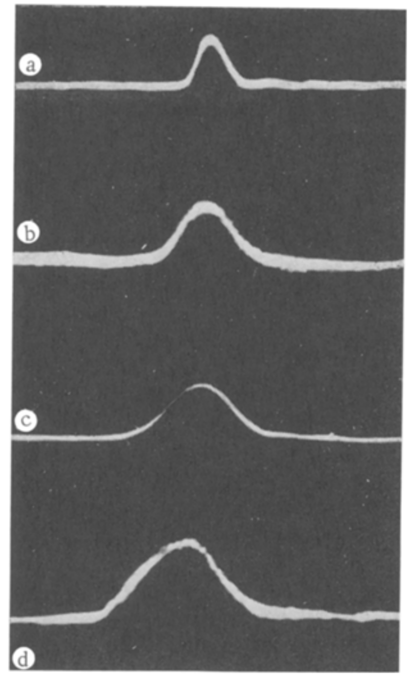

Fig. 6

Fig. 5. The spatial distribution of the laser radiation intensity in the cross section of the beam in the far zone of the exit diaphragm. Distance from diaphragm — 3 m, spot diameter — 4 mm.

Fig. 6. Oscillograms of output radiation of ruby laser at various pulse durations: a) $\tau = 1.4$, nsec; b) $\tau = 2.5$ nsec, c) $\tau = 3.5$ nsec; d) $\tau = 5$ nsec.

The shortest radiation pulse duration, as noted above, was determined by the slope of the pulse of the control voltage and was limited in our laser system by the temporal characteristics of the elements of the electric supply circuit for the Pockels cell. To obtain shorter radiation pulses ($\tau < 1$ nsec) it is necessary to use a discharger and other shutter-supply electric circuit elements with improved temporal characteristics. It appears that the difficulties here lie only in the construction. In particular, we succeeded in obtaining radiation pulses even shorter than 1 nsec ($\tau \sim 0.5$ nsec). The output power of the radiation was practically independent of the pulse duration at $\tau < 2L/c$ and reached $\sim$10 MW.

As to the spectrum of the output radiation, it was not directly investigated. There is every ground for assuming, however, that it was single-frequency (the width of the spectrum was equal to the natural width determined by the duration of the radiation pulse), inasmuch as the master laser operated in the regime with a single longitudinal mode since a quartz stack was the selector.

It should be noted that the characteristics of the output radiation were stable under variation of the generator and amplifier pump power, and its intensity could be varied in a wide range by varying the pump power. One of the advantages of the described system is that it is not sensitive to optical inhomogeneity of the active elements: we used regular commercial ruby crystals in this installation. Yet it is known that the production of single-mode radiation in standard fashion by the mode selection inside the resonator is very sensitive to the quality of the crystals and to the excess of the pump power over threshold.

We note that the principle proposed and realized in the present study for shaping a spatially homogeneous radiation field and the equivalent radiation of a single-mode laser by spatial filtration of the radiation by a system of diaphragms can be used to develop high-power laser systems of various types (solid-state and gas lasers), operating both in the millisecond and in the cw regimes, where the problem of obtaining single-mode radiation is even more difficult than in the case of short laser pulses.

Procedures Used in the Investigation of Laser Damage

As indicated above, success in the study and in the understanding of the mechanism in processes of laser damage to transparent materials is determined to a considerable degree by the procedure used. In the earlier investigations of damage, the most common methods made it possible merely to establish the onset of damage at a threshold radiation intensity by the appearance of a spark at the instant of breakdown or by visible mechanical damage to the material — symptoms that provide little information. Numerous attempts were undertaken later on to investigate different dynamic effects that develop during the time of passage of the damaging laser pulse. A study of these dynamic effects yields the most complete information on the mechanism and dynamics of the laser damage.

These methods include an investigation of the optical emission accompanying the material damage, and various characteristics of this emission were investigated by many workers. Thus, the emission spectrum was measured (see, e.g., [16, 99]) as well as its time evolution [100, 101]. A study was also made of the propagation of the luminous plasma from the surface of the dielectric in the course of damage [9]. The result yielded the plasma temperature and an estimate of the recombination time of ions in the plasma [101]. It was also established that in the case of surface damage to a dielectric the plasma is more accurately seen as a consequence [9] than the cause of the damage, in contrast to earlier assumptions. The last result can be seen to have a bearing not only on the stage of development of the damage, but also on the mechanism whereby the energy is absorbed from the laser beam and damage is produced.

The compression waves produced at the points of damage were observed using both shadow [16] and holographic procedures [17], which made it possible to measure the velocity of such waves and to conclude that the greater part of the absorbed laser beam energy is transformed into the energy of elastic waves. Additional illumination of the damage region from the side with an He—Ne laser [7] made it possible to study the dynamics of the damage and to establish that it develops mainly after the passage of the laser pulse.

Additional information is obtained from investigations of the distortions of the transmitted light pulse during the damage. For example, from the value of the dip it is possible to assess that part of the radiation which was trapped in self-focusing [44]. Observations of the character of the cutoff of the transmitted pulse have made it possible in the opinion of Bass and Fradin [63] to separate the cases of damage to the material proper from damage at the inclusions.

A similar conclusion concerning the damage mechanism was made in investigations [48, 49, 57] of the morphology of the resultant damage. It should be noted that the effect of self-focusing in solids is frequently evaluated also from the morphology of the damage (the transverse dimension of the damage track is in this case much smaller than the dimension of the caustic of the focusing lens).

In a number of studies [99, 100], the method used to investigate the laser damage was to study certain aspects of light scattering in the course of damage. The intensity of the forward light scattering increases strongly and amounts to ~ 0.01 of the intensity I_0 of the incident light, and after the passage of the damaging pulse the scattering increases to $0.1I_0$. The latter fact indicates that the damage develops mainly after passage of the laser pulse. This conclusion, however, is in essence the only consequence of the study of the scattering of light in connection with the damage problem, although the light scattering procedure is capable of yielding, in principle, much more information on the damage-inducing processes of interaction of radiation with matter.

So far, the connection between light scattering in transparent materials and material damage under the influence of high-power laser radiation has not received the attention it merits. We have therefore investigated this question and have developed new procedures for obtaining laser damage by using various characteristics of light scattering.

The study of light scattering at low intensities has enabled us, on the basis of the proposed purity criterion, to determine the optical quality of the crystals and glasses which served as objects for the investigation of the laser radiation. This question was treated in greater detail in Sec. 2.

Examination of light scattering by absorbing inhomogeneities of the medium at high intensities, carried out in [102, 103], has led to discovery of nonlinear scattering, which was observed in corundum crystals [78] at intensities close to the damage threshold. Subsequently, the nonlinear scattering of the light was investigated also in other materials; it turned out that it is either observed at intensities somewhat lower than the damage threshold or accompanies the damage process. This indicates that the mechanisms of interaction of radiation with matter, on which both phenomena are based, are related, and that it is important to investigate them jointly. Thus, nonlinear scattering of light (which precedes or accompanies the damage process) characterizes the initial stages of the mechanisms of interaction of laser radiation with matter which lead to damage, and it is one of the principal methods of investigation of these interactions.

Scattering of light at intensities close to the damage threshold (both below and above it) was investigated at an angle of 90° to the propagation direction of the incident radiation with the aid of an ÉLU-FT electron beam multiplier and an I2-7 oscilloscope, using the laser setup illustrated in Fig. 4. The intensity of the incident light was increased by transferring the neutral light filters from position 6 to position 7, so that under conditions when the scattering was linear in the radiation intensity the same amount of light was incident on the photoreceiver. Elimination of the influence of the instability of the laser output power was done with the aid of an optical delay line, which made it possible to carry out a comparison of the intensities of the incident and scattered radiation. All measurements of the amplitude or waveform of the scattering pulse observed by this procedure point to a nonlinear character of the scattering, which can be both reversible (not connected with the formation of mechanical damage in the medium and thus constituting a nonlinear effect, in the sense understood in nonlinear optics) and irreversible (with formation of mechanical damage in the dielectric). The latter is established by observing the light scattering at intensities much lower than those used to observe nonlinear effects in the scattering. The return of the scattering pulse to its initial form means reversibility; in the opposite case we have irreversibility. Results of a detailed investigation of light scattering near the damage threshold in various transparent dielectrics will be presented in Sec. 5.

Closely connected with the question of the nonlinear character of the scattering of light is the crossed-polaroid procedure, which also yields information on the initial stages of the damage process. The gist of this procedure is that one polaroid is placed in front of the investigated object (a Glan prism was used in our experiments), and this transmits the principal polarization of the incident radiation. A second (crossed) polaroid, which does not transmit the principal polarization of the incident radiation, is placed in front of the recording system. The method of crossed polaroids offers additional advantages over the method of nonlinear light scattering. These advantages are connected with the fact that changes in the scattering are registered in this case against a background of much lower intensities (by more than three orders of magnitude under our experimental conditions) than those registered when nonlinear scattering of light is observed. Therefore the sensitivity of this method to nonlinear changes in the scattering intensity is much higher, meaning that it makes it possible to investigate more effectively the initial predamage stages of interaction of the laser radiation with the matter. The crossed-polaroid procedure also makes it possible to record changes in the intensity of the light passing through the sample; these reflect the process of absorption of energy from the laser radiation and its subsequent transformations.

One of the advantages of the method of nonlinear scattering of light with crossed polaroids is that the latter make it possible to fix the changes of the scattering intensity due to changes in the refractive index of the medium precisely in the interaction region, something difficult to do when using probing radiation. To be sure, in this case these changes are registered only over times equal to the duration of the laser pulse, and the entire dynamics of the damage cannot be tracked.

Nonlinear scattering of light with crossed polaroids was used in the investigation of the damage dynamics. These investigations were made with a laser that produced giant pulses with 20-nsec duration (see Fig. 4). The results of investigations with different dielectrics will be presented below.

The methods considered above make it possible to assess the entire range of interaction as a whole, without providing a spatial picture of the evolving process. Of particular interest is an investigation of local scattering of light in the damage region with the aid of a microscope. The gist of such an experiment is that the caustic of the lens that focuses the incident radiation into the interior of the investigated sample must be in the field of view of the microscope objective; light scattered from the caustic region may be registered either visually or on photographic film. This method yields information on the scattering of light by individual centers and on the change in light intensity due to local changes in the refractive index. A similar procedure was used in the study of light scattering at low intensities by the ultramicroscopy method [81]. However, this procedure has not been used so far to investigate light scattering at intensities close to the damage threshold. The proposed procedure was used to investigate damage in crystals and glasses with the aid of a single-frequency ruby laser with variable pulse duration in the nanosecond band. The results are given in Sec. 5.

Experiments on laser damage using the procedures described above were performed in the following manner. The laser radiation was focused on some point inside the sample at a power lower than the damage threshold. The presence or absence of damage in the sample was verified with the aid of a He–Ne laser whose radiation was guided to the same point of the sample by focusing with the same lens as for the principal radiation. If no damage was observed, then the ruby-laser radiation was increased by $\simeq 15\%$ and a new flash was produced at the same point, followed with a check on the absence of damage in the sample. This procedure was continued until visible damage was observed, which was always accompanied by high-temperature glow (spark). The sample was then shifted to a new position and the investigation repeated. Usually this method was used to perform 10-20 measurements of the damage threshold at each laser pulse duration. The threshold was taken to be the maximum peak laser radiation that could pass through the sample without producing damage.

5. INVESTIGATIONS OF THE MECHANISM OF LASER DAMAGE IN CRYSTALS AND GLASSES

In this section we present the results of the investigation of various aspects of the problem of laser damage to transparent materials: 1) the influence of various types of impurities and of their valent and structure-phase state on damage to ruby crystals; 2) determination of the damage mechanism in sufficiently pure crystals and glasses and the associated determination of the role of minute absorbing defects which are always present in real dielectrics; 3) study of the dynamics of damage in crystals and glasses.

The importance of these aspects in the damage problem is dictated by the following considerations. Whereas the laser damage produced in glasses by absorbing inclusions was investigated in a number of studies, and while certain distinguishing features of the damage (in particular, the role of large platinum inclusions) have been made clear, the influence of various impurities and defects in crystals on the damage process has hardly been investigated (we mention only one study [41] in which the influence of certain defects on the damage threshold was noted). This question has been therefore considered in detail with ruby crystals, which are of practical importance in laser technology, chosen as the example.

The question of the decisive mechanism of volume damage in sufficiently pure transparent materials remained open until recently. The authors of a number of papers that were examined in detail in the survey of the literature on damage have stated that the mechanism of laser damage in crystals that do not contain large absorbing inclusions is an electron avalanche that develops in a pure defect-free matrix, but this conclusion, as shown in the survey of work on laser damage, has not been adequately established. We therefore investigated the mechanism of laser damage in sufficiently pure crystals and glasses.

Influence of Various Impurities on the Laser Damage of Ruby Crystals*

Many papers devoted to the damage of ruby crystals by laser radiation have by now been published. However, the question of the decisive mechanism of volume damage remains debatable. In particular, the explanation that the damage mechanism is avalanche ionization of the crystal matrix can hardly be regarded as fully established, since there is an appreciable (by up to 100 times) spread in the experimental values of the threshold [21]. This spread seems to point to an important role of defects in the mechanism of laser damage of ruby, something not seriously considered in the literature so far. We have therefore

*This part of the work was performed jointly with the Crystallography Institute of the Academy of Sciences of the USSR with the help of V. Ya. Khaimov–Mal'kov and the members of his group.

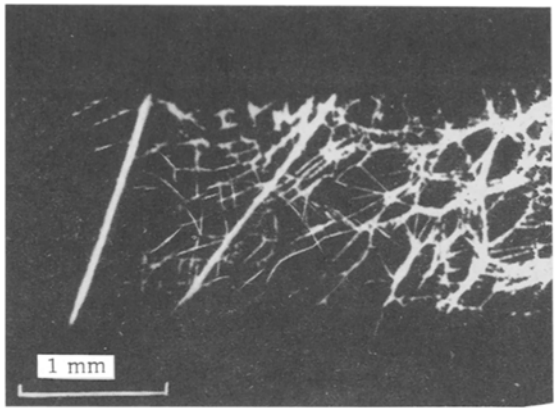

Fig. 7. Distribution of nickel inclusions in a sapphire crystal as observed in an ultramicroscope.

undertaken a study of the influence of various impurities on the threshold of volume destruction of ruby crystals in the giant-pulse regime.

We investigated ruby and sapphire crystals grown by the Verneuil method, in which titanium, vanadium, iron, cobalt, nickel, and magnesium impurities were introduced. The concentration of the impurities in the samples was 10^{-3}-10^{-4}%.* These elements were chosen because they usually are present in ruby crystals used in lasers. Depending on the conditions of the growth and subsequent heating of the crystals, these impurities can change the valence and produce foreign phases (inclusions).

The change in the valence state of the impurities was effected by high-temperature heating (to $T \simeq 1800°C$) of the crystals. The valence state of the impurities that enter the lattice isomorphically was determined from the optical absorption and luminescence spectra. The method of ultramicroscopy in scattered light was also used to determine the presence of foreign inclusions that could be produced by the impurities introduced into the crystals. This method made it possible to detect the presence in the crystals of individual particles with dimension $a \gtrsim 3 \cdot 10^{-6}$ cm.

It was observed that in sapphire samples, after heating in vacuo, the titanium impurity is in the Ti^{3+} state and it replaced the Al^{3+} ions isomorphically. If the heating is in an oxygen atmosphere, the titanium goes over into the Ti^{4+} state. Some of the ions then make up a foreign phase whose rate of precipitation increases sharply with increasing temperature. The structure of this phase remains unclear, but it can only be assumed that it constitutes inclusions of either TiO_2 or of aluminum titanate.

Impurities of nickel, cobalt, and iron, after the sapphire samples are heated in an oxygen atmosphere, form trivalent ions that replace Al^{3+} isomorphically. Heating of the same samples in vacuo leads to the transformation of the ion into the divalent state. Some of the ions enter the sapphire lattice isomorphically, and some are precipitated into a foreign phase. Investigations of the distribution of this phase by the ultramicroscopy method have shown that the precipitation of the inclusions takes place mainly on block boundaries, dislocations, and vacancy clusters (Fig. 7). A microscopic x-ray phase analysis of the precipitated particles has shown that the reduction of the Ni, Co, and Fe impurities can proceed up to the metallic state.

In samples with a magnesium impurity, heating in an oxygen atmosphere produces a foreign phase in the form of small inclusions that vanish after subsequent heating in vacuo.

In sapphire crystals with a vanadium impurity, no inclusions of a foreign phase are produced, and only the valence transitions $V^{2+} \rightarrow V^{3+}$ take place when annealing is done in an oxygen atmosphere rather than in vacuo.

*We note that these concentrations do not exceed the content of the uncontrollable impurities by more than one order of magnitude, and were chosen only to clearly reveal the influence of specific impurities in the damage mechanism.

TABLE 2

Annealing atmosphere	Impurity					
	Ti	V	Co	Ni	Fe	Mg
Oxygen	$3\cdot10^{-2}$	$5\cdot10^{-2}$	0,5	0,6	0,6	$7\cdot10^{-2}$
Vacuum	0,8	$7\cdot10^{-2}$	10^{-2}	$2\cdot10^{-2}$	10^{-2}	0,9

We note that the valence transitions of the impurities and the precipitation of the foreign phase are fully reversible when annealing atmosphere is changed.

Volume damage of sapphire and ruby samples containing the indicated impurities was investigated under the influence of giant ruby laser pulses ($\tau \simeq 40$ nsec) using the setup described above. Measurement of the damage thresholds P_d of samples with different impurities was made relative to the threshold P_d° of an impurity-free control sample of sapphire at different annealing temperatures (Table 2), with the laser operating in the multimode regime. An estimate of the damage threshold of the control sample under the influence of a single-mode laser radiation yielded a value of $\simeq 2\cdot10^{10}$ W/cm^2.

An analysis of the results shows that in those cases when a foreign phase is precipitated in the ruby and sapphire crystals, an abrupt decrease of the threshold of volume damage (by up to 50 times) is observed. In the absence of foreign inclusions, the values of P_d of all the samples investigated, both sapphire and ruby, do not differ significantly, despite the differences in the impurities introduced and in their valence states. The apparent cause of the last fact is that the crystals contain small ($a < 3\cdot10^{-6}$ cm) inclusions of foreign phase, produced by uncontrollable or introduced impurities, which could not be resolved by the untramicroscope used by us. Evidence of the presence of such inclusions is the noticeable Rayleigh scattering, the intensity of which in the investigated samples greatly exceeded the intensity of the Mandel'shtam–Brillouin scattering in crystalline quartz (in sapphire with Ni impurities we have $\eta = 17.0$ (see Table 1)).

The foregoing data point to a substantial role for the absorbing defects in the mechanism of laser damage of ruby crystals. The decisive damage mechanism in real crystals is apparently connected with the presence of impurities. It should be noted that the problem of inclusions is more serious in crystals than in glasses, owing to the low solubility of many impurities in crystals as well as the presence of structural defects in them that facilitate the formation of foreign phase inclusions.

Role of Absorbing Defects in the Mechanism of Laser Damage to Real Transparent Dielectrics

1. The authors of a number of recent papers [48, 49] believe that they have succeeded in observing damage in a number of crystals and glasses without the influence of absorbing inclusions or self-focusing. The results of the experiments were interpreted by the authors on the basis of an electron avalanche that develops in the pure material. As indicated above, however, this assumption is questionable. Consequently, the question of the dominant mechanism of laser damage in sufficiently pure optical materials (i.e., those not containing large absorbing inclusions) remains open.

In this section we examine the mechanism of laser damage of real transparent dielectrics using a number of crystals and glasses as examples. We pay close attention in the examination of this question to a clarification of the criterion of optical purity, the influence of self-focusing, and the role of microdefects in the damage process.

In the determination of the physical mechanisms responsible for damage we are mainly concerned with the 0.1-10-nsec range of laser pulse durations, inasmuch as the characteristic times of establishment of various nonlinear processes can lie in this interval. The experiments were therefore performed at pulse durations of 1-10 nsec.

2. The investigations of the crystal and glass damage were performed using a single-frequency ruby laser with variable pulse duration in the nanosecond range (1.4-7 nsec). The laser had a spatially homogeneous radiation field with diffraction divergence. A detailed description of this laser is given in Sec. 4. A diagram of the laser setup is shown in Fig. 8. In the experiments we registered the threshold powers of the input radiation corre-

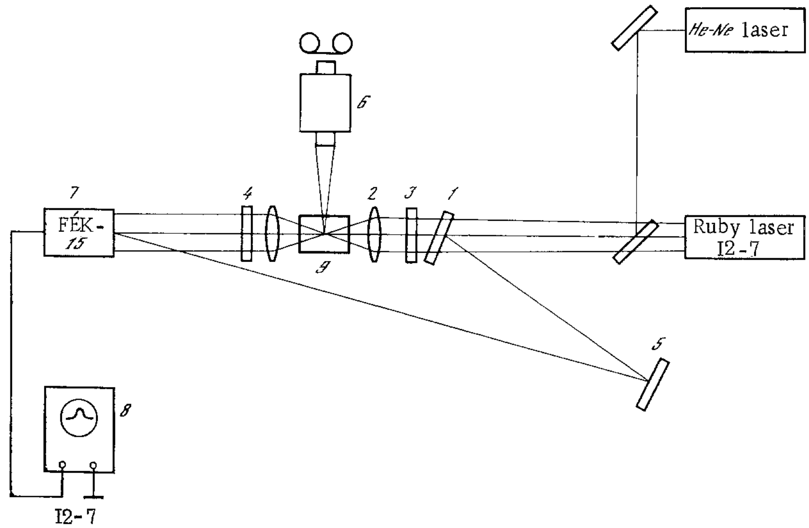

Fig. 8. Diagram of experimental setup for the investigation of laser damage: 1) beam-splitting plate for optical delay of the incident radiation pulse; 2) focusing lens; 3-5) sets of neutral light filters.

sponding to the appearance of damage, the morphology of the damage, the spatial structure of the glow and scattering of the laser radiation in the region of the caustic of the focusing lens, as well as the distortions of the waveform of the transmitted light pulse. The ruby laser radiation was focused on the volume of the investigated samples with the aid of lenses with different focal lengths (F = 18 and 90 mm). A probing He–Ne laser was used to indicate the possible presence of large defects and inclusions in the samples and the onset of the damage. The spatial structure of the glow accompanying the damage and the light scattering of the ruby laser in the region of the caustic of the focusing lens were studied with a microscope. We note that this procedure has not been used before in experiments on laser damage. The procedure turned out to be quite effective and made it possible to study local variations of the medium in detail at the instant of passage of the damaging laser pulse.

3. The results obtained when determining the optical purity of the crystals and glasses suggest that the defects and inclusions present even in very pure optical materials can play an important role in laser damage. We therefore investigated damage in the same samples in which the light scattering was investigated. It turned out that the character of damage in these samples differs significantly and depends on the purity of the optical material.

In sapphire crystals with a nickel impurity, which have a considerable light scattering ability (η = 17.0), the damage took the form of individual "stars" randomly located in the caustic of the focusing lens (this indicates the absence of a substantial influence of self-focusing). The laser damage threshold was much lower here (approximately by one order of magnitude) than in pure sapphire. The character of the damage, which remained the same at different focal lengths of the focusing lens (F = 18 and 90 mm), and the relatively low threshold ($\sim 10^{10}$ W/cm^2) indicate that the damage mechanism is connected with the presence of absorbing defects in the material. An investigation of the dependence of the damage threshold on the duration τ of the laser radiation pulses in the 1.4-7 nsec interval has shown that it does not change with a change of the pulse duration. This makes it possible to estimate, on the basis of the thermal damage mechanism theory developed in Sec. 3, the sizes of the absorbing inclusions responsible for the damage ($a \ll \sqrt{\chi\tau} = 2\cdot 10^{-5}$ cm, where χ is the thermal diffisivity coefficient of sapphire). This estimate agrees with the size estimate based on light scattering data.

Study, with a microscope, of the local scattering of light during the passage of the laser pulse has confirmed our previous conclusion that the damage in sapphire samples with Ni impurity is due to the presence of centers that absorb the laser radiation. We note that

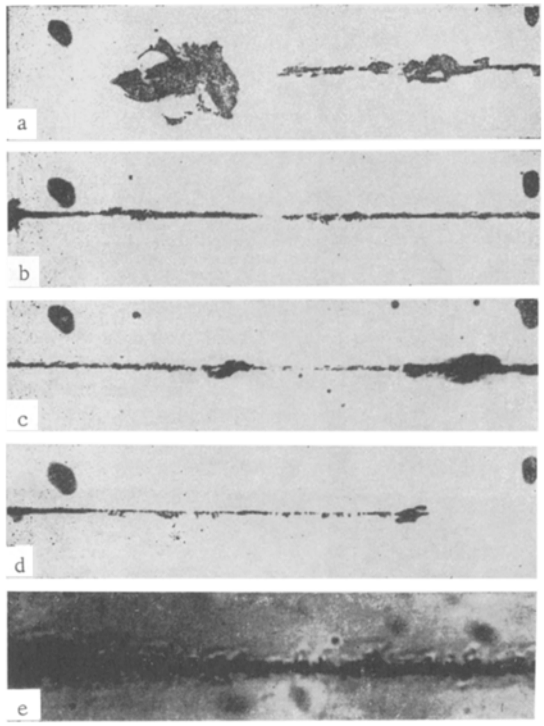

Fig. 9. Morphology of the damage track produced in sapphire by focusing laser radiation with a lens of F = 90 mm: a-d) damage filament (d — scale 100 μm); e) section of damage filament at large magnification (scale 50 μm).

below the damage threshold I_d (at an incident radiation intensity of $I \simeq 0.9\ I_d$) no local scattering of light from the region of the caustic was observed. The comparison of this correlation with the already reported strong scattering at intensities above the threshold indicates that nonlinearly scattered light is observed here.

Thus, the investigation of the laser damage of sapphire crystals containing inclusions with $a < 10^{-5}$ cm has shown that the principal damage mechanism is connected with the presence of such inclusions, and that self-focusing exerts no substantial influence even when the intensity of the incident radiation appreciably exceeds the threshold.

4. The damage to sufficiently pure crystals in glasses, which have weak light scattering, is of an entirely different character. Data on the intensity of light scattering in these samples are given in Table 1.

When the laser radiation is focused with the F = 90 mm lens (the minimum transverse dimension of the caustic was $d_c \simeq 40$ μm), the damage produced in all the optical materials took the form of filaments (diameter $\simeq$1-2 μm and length $\leq$ 1 cm), thus indicating that self-focusing plays the decisive role in the damage process (see Fig. 9). The threshold powers of the radiation in all the crystals and glasses were independent of the laser pulse duration τ in the 1.4-7-nsec range. The exceptions were sapphire crystals, for which the threshold powers were independent of the duration at $\tau \geqslant 2.5$ nsec and whose threshold increased (by approximately 1.5 times) at $\tau = 1.4$ nsec. Table 1 shows the values of the threshold powers of the radiation at $d_c \simeq 40$ μm (the value for sapphire is given at $\tau \geqslant 2.5$ nsec). We note that the true threshold densities of the radiation remain undetermined here, since both the fraction of trapped energy and the cross section of the laser beam in the medium with self-focusing remain unclear.

A detailed study of the morphology of the damage track produced (see Fig. 9) has shown that it has neither a large-scale structure (i.e., damage regions with a length of the order of hundreds of microns or more, between which there are randomly located regions of undamaged material of length ∿100 μm) nor a small-scale structure (where each region of the

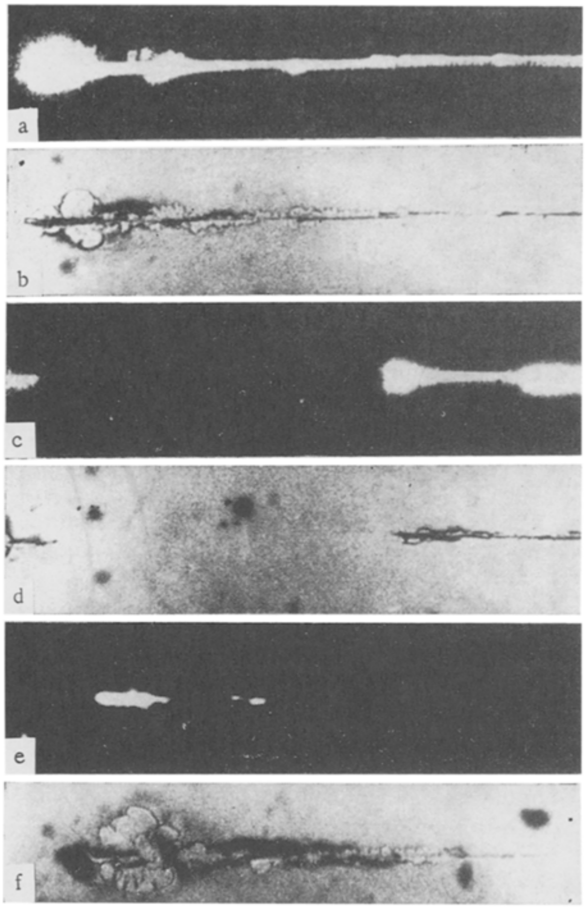

Fig. 10

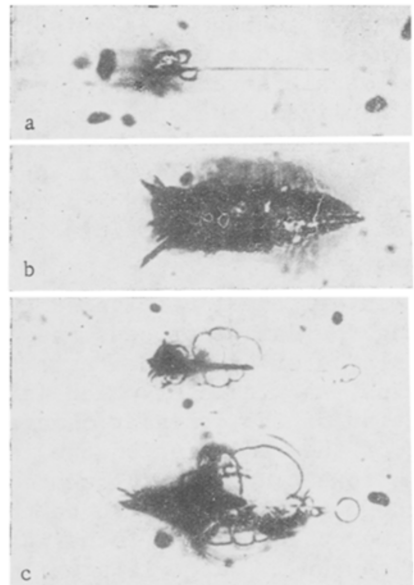

Fig. 11

Fig. 10. Pictures of light scattering and light emission, and the corresponding damage, obtained with a microscope from the region of the caustic of a focusing lens with F = 90 mm: a) scattering in BK-104 glass near the stopping point of the nonlinear focus; b) damage; c) scattering in BK-104 glass from the region corresponding to a break in the damage filament; d) damage; e) light emission in laser glass; f) damage (scale 100 μm).

Fig. 11. Morphology of damage produced when laser radiation is focused with an F = 18 mm lens: a) damage in TF-8 glass; b) damage in crystalline quartz; c) damage in BK-104 glass.

track consists of separate damaged spots with dimensions of approximately 1 μm, separated from one another by several microns and arranged along the track). To determine the causes of formation of such a track structure we investigated the local scattering of light and the light emission from the region of the caustic of the focusing lens during the time of passage of the damaging laser pulse. These investigations have shown that the scattered light contains a large- and small-scale structure that correlates with the morphology of the damage tracks (see Fig. 9). One should note a number of distinguishing features of the corresponding damage regions, revealed by examination of the pictures of the light scattering during the time of the damaging laser pulse. First, near the stopping point of the nonlinear focus there is a light region of considerable length, which apparently points to a considerable increase of the elastic stresses in the medium [104]. Second, the damage track is interrupted in those places at which the light picture shows a region of increased light scattering. Third, Fig. 10a shows spherical regions similar to those observed in the scattering of light in the case of damage to sapphire crystals with Ni impurity (Fig. 9a). Just as in the case of sapphire with Ni, there is a correlation between these regions and the damage centers (Fig. 10b), which indicates that a considerable influence is exerted on the damage process by absorbing defects. We call attention to the comb-like small-scale structure in the light scattering, the explanation of which requires an analysis of the evolution of self-focusing in nonlinear media in the presence of sources of light absorption on the axis of the laser beam. The structure is apparently due to minute absorbing defects which initiate the damage.

Observation, with a microscope, of the light emission from the region has shown that the transverse dimension of the light emission filament is much smaller than the dimensions of the caustic and corresponds to the width of the produced damage track (see Fig. 9).

The results can be easily understood if we consider self-focusing with allowance for the absorbing microdefects. By microdefects we mean here regions of small dimensions with a local coefficient of light absorption (linear or nonlinear) greatly exceeding the absorption coefficient of the surrounding matrix. According to the model of Lugovoi and Prokhorov [6], the formation of damage tracks is the result of motion of a nonlinear focus in the medium. If we take into account the presence of absorbing defects in the medium, then the large- and small-scale structure of the damage track can be explained in the following manner. When the nonlinear focus moves, energy is absorbed by the microdefects which initiate the damage and lead to the formation of a small-scale structure. When the linear focus approaches a larger absorbing defect, strong perturbations of the refractive index are produced in the medium surrounding the defects as a result of thermal heating, and these lead to defocusing of the radiation. This is apparently the cause of formation of breaks in the damage filaments.

5. Since in all the investigated crystals and glasses which have relatively small light scattering the damage process passed through a self-focusing stage, elimination of this effect would be of great interest; this would make it possible to measure the true damage thresholds, knowledge of which is essential when it comes to comparison with theory and to explanation of the dominant damage mechanisms. It was reported in [48, 49] that by sharp focusing of the radiation on the interior of the investigated samples ($d_c \simeq 15$ μm) it was possible to exclude the influence of self-focusing and observe the true damage mechanism, which the authors believe to be the electron avalanche mechanism. A similar experiment was performed using as the focusing lens an 8^x microscope objective (F = 18 mm). The dimension of the caustic was $\simeq 12$ μm. In this case the threshold powers of the radiation entering the investigated sample were decreased in all crystals and glasses by a factor of four compared with the thresholds obtained when the lens with F = 90 mm was used, thus corresponding approximately to the relation $P_d \sim F$. In all the samples, the ratios of the threshold powers and their dependences on the durations of the laser pulses remained the same.

A study of the morphology of the damaged sections using a short-focus lens (F = 18 mm) has shown that in all materials (especially in TF-8 glass) they are filamentary in character (Fig. 11). The ratio of the thresholds in different materials remained the same when the focal length was decreased. Both these facts indicate that in all the considered crystals and glasses self-focusing plays a substantial role in the damage process.*

Investigation of the glow and local scattering of light from the region of the caustic of the F = 18 mm focusing lens yielded qualitatively the same result as in the case of the long-focus lens: 1) The regions of light emission and scattering had a filamentary structure with transverse dimension much smaller than the diameter of the caustic of the lens; 2) the character of the damage was strongly influenced by the absorbing defects (Fig. 12).

A study of the waveform of the pulse of the light passing through the sample at different durations in the range 1.4-7 nsec has shown that when radiation is focused with an F = 18 mm lens there is a rather abrupt cutoff of the pulse on the leading front, which is furthermore at a radiation power less than the peak power in the preceding nondamaging pulse (Fig. 13). This is precisely the reason why the damage threshold was taken to be the maximum peak power of the radiation passing through the sample without producing damage.

Figure 13e shows an oscillogram of the transmitted and incident radiation pulses in the case of damage in sapphire with an Ni impurity. It is seen from the figure that the pulse cutoff is smoother in character. We note that a similar pulse cutoff character was observed in [63], but there the abrupt cutoff was assumed to be evidence in favor of a decisive role of the electron avalanche in the damage mechanism.

*This result casts doubt on the conclusion of the authors of [48, 49] that they excluded the influence of the self-focusing effect from their experiments.

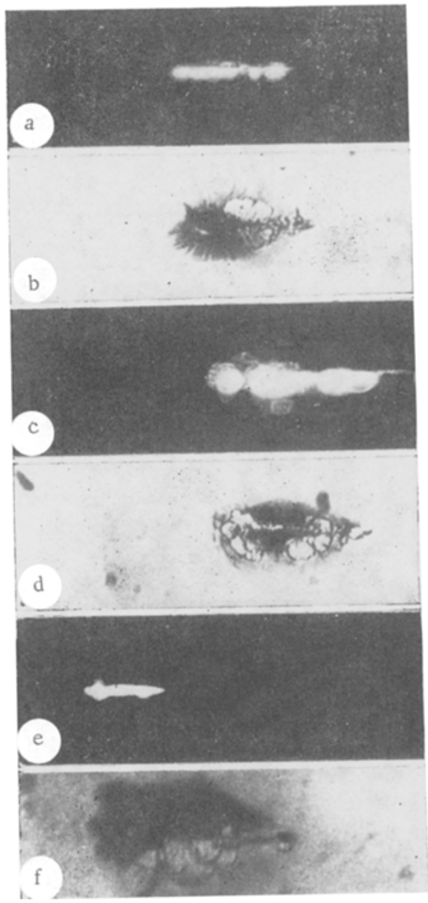

Fig. 12

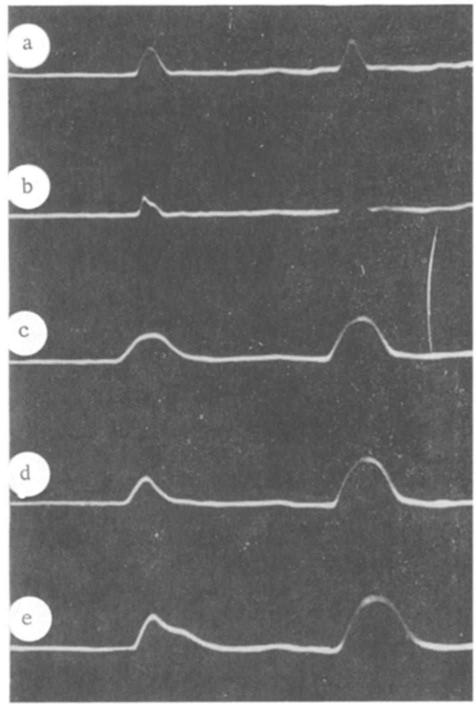

Fig. 13

Fig. 12. Pictures of light scattering, light emission, and corresponding damage, obtained with a microscope from the region of the caustic with an F = 18 mm focusing lens: a, c) scattering in sapphire; b, d) damage; e) light emission in BK-104 glass; f) damage (scale 100 μm).

Fig. 13. Oscillograms of pulses of the transmitted and incident radiation at powers close to the damage threshold in sapphire, using a short-focus lens (F = 18 mm) and different laser pulse durations: a, c) below the threshold; b, d) above the threshold of damage in pure sapphire; e) above the threshold of damage in sapphire with an Ni impurity.

Thus, the entire set of results on damage in sufficiently pure crystals and glasses, using a short-focus lens, shows that by this method it is impossible to exclude the influence of self-focusing, which is the limiting factor that determines the optical endurance of the materials. The investigations performed show also that in the course of damage an important role is played by absorbing defects. These results indicate that the question of the true mechanisms of laser damage which determine the limiting endurance of transparent dielectrics remains open. In any case, in the theoretical analysis of these mechanisms it is necessary, in our opinion, to take into account the presence of absorbing defects, which apparently are always contained in real media.

Dynamics of Laser Damage of Crystals and Glasses.

Accumulation Effect

In Sec. 3 we developed a nonlinear theory of the thermal mechanism of damage, within which the threshold conditions can be found without considering the dynamics of the thermoelastic stresses that lead to mechanical damage of the material, since in this theory the

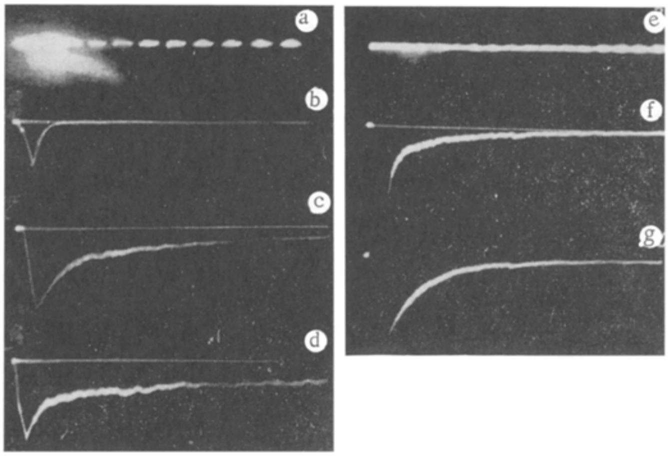

Fig. 14

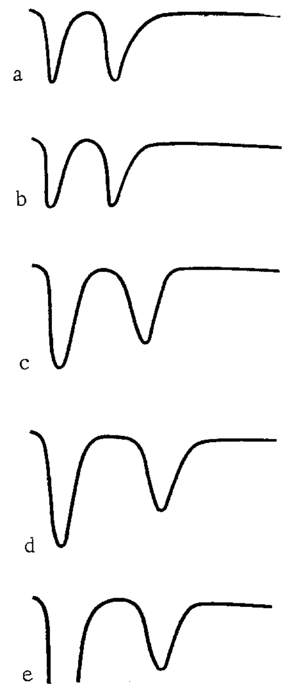

Fig. 15

Fig. 14. Oscillograms of optical emission pulses in volume destruction of crystals and glasses: a) 20-nsec markers; b) laser radiation pulse; c) emission in TF-8 glass; d) emission in laser glass; e) 50-nsec markers; f) emission in TF-8 glass; g) emission in sapphire.

Fig. 15. Oscillograms of scattered-light pulses in BK-104 glass and of the incident radiation of intensity I near the threshold of laser damage I_d. a) $I = 0.1 \cdot I_d$; b) $I = 0.8 \cdot I_d$; c) $I = 0.1 \cdot I_d$; d) $I = 0.8 \cdot I_d$ — damaging pulse; e) $I = 0.1 \cdot I_d$ — scattering by the damage region.

damage mechanism has the features of thermal explosion. However, as indicated above, a complete description of optical damage must include both the mechanism of absorption of the laser radiation energy and the dynamics of the damage process; the latter can be traced by studying the kinetics of light scattering by the damage region and the scattering of the accompanying optical emission. It is therefore very important to employ techniques of light scattering investigation at intensities close to the damage threshold. Such investigations can yield valuable information on the dynamics of the laser damage, inasmuch as the scattering and the damage may have a common cause.

The study of light scattering and light emission in the course of damage has been the subject of a large number of papers (see, e.g., [100]). It has been established that the damage develops much later than the passage of the laser pulse, and the forward scattering intensity amounts to $\sim$(0.01-0.03) of the threshold radiation intensity I_d during the time of the pulse, and to $\sim 0.I_d$ after the passage of the light pulse. The most detailed investigation of the kinetics of the scattering and optical emission in damage of alkali-halide crystals, sapphire, and laser glass is reported in [99], where it was found that on passage of a damaging laser pulse of duration 10 nsec an increase in the scattering is observed after 3-5 nsec, the scattering is polarized, and its intensity is $\sim 0.01\ I_d$. It was found that the threshold of the optical emission coincides with I_d, the entire emission spectrum is produced at one and the same time, and its intensity is $\simeq 0.1$ of the intensity of the scattered light.

We have investigated the optical emission that accompanies the damage in sapphire crystals, TF-8 glass, BK-104 glass, and laser glasses. It turned out that the character of the emission is significantly different in the cases of damage of crystals and glasses (Fig. 14). At a laser pulse duration of $\simeq 20$ nsec, the emission in the sapphire crystal has a characteristic attenuation time $\simeq 100$ nsec. Figure 14 shows the emission pulses in the case of volume damage in glasses, and a noticeable difference from the preceding case is observed:

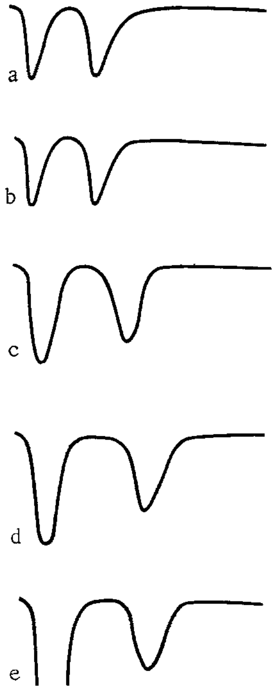

Fig. 16

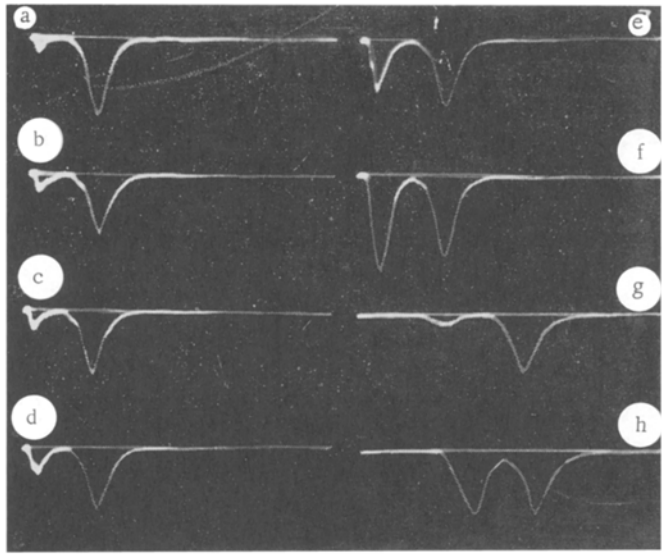

Fig. 17

Fig. 16. Oscillograms of successive pulses of scattered light in TF-8 glass and of the incident radiation. a) $I = 0.1 \cdot I_d$; b) $I = 0.6 \cdot I_d$; c) $I = 0.8 \cdot I_d$; d) $I = 0.8 \cdot I_d$ — damaging pulse; e) $I = 0.1 \cdot I_d$ — scattering by the damage region.

Fig. 17. Oscillograms of successive pulses of incident radiation and of scattered light in crossed polaroids in TF-8 glass (a-d) and in BK-104 glass (g, h). a) $I = 0.1 \cdot I_d$; b) $I = 0.6 \cdot I_d$; c) $I = 0.1 \cdot I_d$; d) $I = 0.6 \cdot I_d$; e) $I = 0.6 \cdot I_d$ — damaging pulse; f) $I = 0.1 \cdot I_d$ — scattering by the damage region; g) $I = 0.8 \cdot I_d$; h) $I = 1.0 \cdot I_d$ — damaging pulse (intensity of scattered light in the nonprincipal polarization increased by more than 20 times).

the pulse has a clearly pronounced biexponential character.* The point of transition from one exponential to another on the optical emission pulse in glasses corresponds approximately to the end of the laser pulse. This difference between the light-emission kinetics in crystals and glasses is apparently due to the difference in the mechanisms whereby energy is carried away from the produced plasma.

As indicated above, valuable information on the dynamics of laser damage can be obtained by investigating the scattering of light by the damage region. The scattering of light at radiation intensities close to the damage threshold (both below and above) was therefore considered. In contrast to [99, 100], where the light scattering was investigated in connection with damage of transparent materials, study of the scattering at intensities lower than the damage threshold makes it possible to assess the prebreakdown state of the substance in processes that lead to the nonlinear character of the scattering and to damage of the dielectrics.

The light scattering in sapphire crystals, BK-104 glass, TF-8 glass, and laser glasses was investigated with the laser setup shown in Fig. 8, using the technique of nonlinear scattering of light with crossed polaroids, as described above. As a result it was established that the character of the changes in the scattering of light depends considerably on the type of material. In sapphire, BK-104 glass, and in laser glasses, these changes were quite abrupt and were observed only in a very narrow interval of radiation intensities near the damage threshold ($I \leqslant I_d$). Figure 15 shows oscillograms of the pulses of the scattered light in BK-104 glass and of incident radiation I in a series of successive laser

*A similar optical emission character was observed in [101] in damage of dielectric surfaces.

flashes. It is seen that near I_d reversible nonlinear scattering of light is observed, but in the case of a slight increase in I a considerable increase of the intensity of the scattered light is observed, as well as a distortion of the pulse waveform, thus indicating that the scattering is nonlinear (Fig. 15d) during the time of the damaging pulse. After formation of the visible damage, a still greater increase of the scattering intensity is observed at $I \ll I_d$, indicating that the damage develops mainly after the passage of the laser pulse.

In TF-8 glass, the changes in the light scattering took place in a larger intensity interval, lower than the material damage threshold I_d, with a single flash ($I > 0.6\ I_d$); they were smoother in character. The light scattering process was reversible under the conditions of the nonlinear scattering technique in the laser radiation intensity interval $0.6\ I_d < I < I_d$ for a series of flashes in the same region of the material; the series was larger the smaller the intensity of the incident radiation. However, following additional shots at the same point, irreversible accumulating changes in the light scattering are observed, and after a certain number of flashes there is produced visible damage to the material, the intensity of scattering from which increases considerably (Fig. 16). This indicates that the damage develops mainly after the passage of the damaging laser pulse.

The accumulation effect in the scattering was even more strongly pronounced when crossed polaroids were used for the observation (Fig. 17). This indicates that the crossed polaroid technique is even more sensitive than the use of principal polarization for the observation of prethreshold irreversible changes in matter subjected to laser action.

The foregoing data can be apparently explained by starting from the assumption that the investigated materials contain microdefects that serve as sources of light scattering and of local microdamage. The observed accumulation effect in scattering and damage then become due to the development (spreading) of the initial defect structure of the investigated materials following repeated action of laser radiation.* As a result, this process of microdamage accumulation leads to formation of a visible macrodamage.

The observed accumulation effect is difficult to explain on the basis of "pure" damage mechanisms (i.e., mechanisms of damage in a defect-free matrix). We therefore do not regard as quite convincing the interpretation of the statistics of laser damage based on the electron avalanche mechanism [64, 65], since it does not take into account the possible accumulation effect. This is all the more so since the statistics observed in [68, 65] were regarded by the authors as one of the main arguments favoring the mechanism of electron avalanche in a pure matrix. We, on the other hand, believe that the presence or absence of the assumulation effect can be regarded as an indicator of the influence of defects on the laser damage.

Thus, the method proposed in the present paper of investigating laser radiation by using nonlinear scattering of light reveals irreversible changes in the matrix of the investigated materials even before the threshold of the visible macrodamage is reached. The effect of accumulation of microdamages, which leads to formation of macrodamage, can be used as a test of the influence of defects on the laser damage on the surfaces and in the volumes of transparent optical materials [105]. An investigation of the scattering of light at intensities close to the damage threshold makes it possible to trace the dynamics of the laser damage, and to explain the processes that lead to the nonlinear character of the light scattering and to the damage.

CONCLUSIONS

We have investigated depth laser damage and light scattering in crystals and glasses. Our investigations have shown a close connection between these two phenomena and made it possible to explain the specific features of the damage processes in real transparent materials. The main results of this paper are the following.

1. We have proposed and investigated a criterion for optical purity of transparent materials as pertaining to the problem of their laser damage. This criterion is based on a comparison of the intensities of the Rayleigh and Mandel'shtam–Brillouin scattering of

*We note that we are speaking here of very pure and optically perfect materials, which, as shown above, nevertheless contain a rather large amount of scattering defects of small (submicron) size.

light at low incident radiation intensity. This criterion was shown to be the most representative test of the purity of the investigated materials from the point of view of the presence in them of various types of defects and inclusions.

2. We developed a nonlinear theory of the thermal mechanism of laser damage with account taken of the dependence of the coefficient of light absorption and of other parameters of the medium on the temperature. This theory explains well the experimental data on laser damage of crystals and glasses containing absorbing inclusions.

3. It was established that the damage to real transparent dielectrics is closely connected with the presence in them of absorbing defects. It was shown that in the purest crystals and glasses the limiting factor is the self-focusing effect.

4. The effect of accumulation in crystals and glasses was observed at intensities close to the threshold of their laser damage and was interpreted as the process of development (spreading) of microdamages initiated by the initial absorbing defects, up to formation of a visible microdamage.

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