NONLINEAR LOSSES IN GENERATORS AND AMPLIFIERS OF ULTRASHORT LIGHT PULSES

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Nonlinear losses were observed in generators and amplifiers for ultrashort light pulses (USP), connected with self-focusing of the laser radiation in the active medium and in other optical media contained in the laser setup. The influence of the nonlinear losses on the dynamics of the development of pulses in the USP generator and on the operation of the USP amplifier is investigated.

INTRODUCTION

AN important problem in the development of powerful ultrashort pulses (USP) in lasers is a determination of the factors that limit the growth of the USP power during the course of amplification. Owing to their high intensity, when USP propagates through the amplifying medium, and also through other optical media contained in the laser setup, one can expect various nonlinear effects to occur and to lead to losses that depend on the power of the amplified radiation. Thus, in^[1] we observed self-focusing of USP in neodymium glass of a high-power laser installation. It is reasonable to assume that the non-linear losses can occur also in generators of USP with mode locking. The resultant losses may turn out to influence strongly the dynamics of the development and formation of the USP. According to the theoretical notions developed $in^{[2-4]}$, USP formed in generators from fluctuations of the intensity of the multimode radiation as the latter passes many times through a medium consisting of an amplifier and a nonlinear absorber with rapid relaxation of the bleached state. Estimates show that when the transverse modes are eliminated, there is a high probability of appearance of a sequence of single USP at the output of the generator. In fact, a sequence of several USP appears much more frequently than predicted by the theory. This fact can be explained by assuming the presence of additional power-dependent losses in the generator. Such losses, which limit the power of the most intense pulses, hinder the formation of a deeplyintensity-modulated sequence of single USP.

There are also other known facts concerning the USP generator, which can be explained by means of the nonlinear-loss mechanism. Thus, it was observed in^[5] that radiation that consists of single USP at the start of the generation train turns out to consist of several pulses at the end of the train. Analogous phenomena were observed by us in an investigation of a ring amplifier for USP, described in^[6]. In^[7] it is reported that a decrease of the thickness of the saturable-dye cell contributes to a more frequent appearance of single pulses. In observations of the multiphoton ef-

fect²⁾ an of stimulated Raman scattering^[8] under the action of a USP generator, it turned out that generator pulses having equal amplitudes on the oscilloscope screen at the beginning and the end of the USP train act differently, namely, the action of the pulses at the beginning of the train is more effective. All these facts can be explained if there exists a nonlinear-loss mechanism that leads to a deterioration of the temporal structure of the generator pulses.

The present investigation was undertaken to ascertain whether there exist nonlinear losses in neodymiumglass USP generators and amplifiers, and if they exist, to investigate their influence on the generators and amplifiers and to determine their nature.

EXPERIMENTAL INVESTIGATIONS OF NONLINEAR LOSSES IN USP GENERATORS AND AMPLIFIERS

The experiment was organized in accordance with the scheme shown in Fig. 1. A diaphragm 1 was placed in the resonator of the USP generator to eliminate the non-axial modes. Q-switching and mode synchronization were with the aid of a cell 2 containing a solution of dye No. 3955 in nitrobenzene. The thickness of the nitrobenzene layer was 1 mm, and the initial transmission of the dye solution at 1.06 μ was 65%. One of the walls of the cell was a mirror with an approximate reflection coefficient 100%, and the reflection coefficient of the output mirror was 70%. Telescope 3 made it possible, first, to compensate for the thermal distortion of the rods during the pumping time and, second, to place in different sections of the beam various materials in which nonlinear losses were to be ob-



FIG. 1. Experimental setup for the study of nonlinear losses in USP generators: 1-diaphragm for eliminating the non-axial modes, 2-cell with nonlinear absorber, 3-telescope, 4-cell with investigated liquid, 5-rods of neodymium glass measuring 10×160 mm, 6-output mirror, 7-coaxial photocell, 8-oscilloscope.

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FIG. 2. Dependence of output energy of USP generator (W-in arbitrary units) on the ratio S of the cross section of the beam in the cell 4 to the cross section of the beam in the active medium for two layers of nitrobenzene: \bullet -3 mm, O-5 mm.



FIG. 3. Diagram of experiment for the study of nonlinear losses in nitrobenzene: 1-neutral light filters, 2-diaphragm, 3-cell with 7-cm nitrobenzene layer, 4-calorimeter.

served. The occurrence of such losses was revealed by the decrease of the output energy of the generator. In the experiment we measured the total energy of the USP train with the aid of an FÉK-15 photocell, an integrating network, and an S1-29 oscilloscope. By placing the cell with the investigated liquid in different sections of the beam inside the resonator it was possible to plot (in relative units) the dependence of the output energy on the section of the beam passing through the cell.

Figure 2 shows such plots for layers of nitrobenzene 3 and 5 mm thick. One can see from the two curves that, starting with a definite intensity (for a specified layer thickness), losses arise in the resonator and increase strongly with increasing intensity. For the thicker layer, these losses arise at lower intensities. This procedure was used to investigate other substances: chlorbenzene, methyl alcohol, and also an empty cell with wall thickness 1 cm. In all these substances we were able to observe nonlinear losses. Filament-like faults were observed in the glass of this cell, and can be naturally attributed to self-focusing.

Investigations of the temporal structure of the radiation has shown that as the additional cell 4 with the nitrobenzene is moved in the direction of lower beam cross sections the operation of the USP generator becomes worse. A large number of additional pulses appeared in the axial period. When the cell was placed in a sufficiently small section of the beam, no sequences of individual spikes were observed at all.

The data indicate that nonlinear losses that depend on the intensity of the flux inside the resonator and on the thickness of the layer of material can arise in the optical materials making up the USP generator.

For a more detailed investigation of the mechanism of the nonlinear losses in nitrobenzene, an experiment was performed in accordance with the scheme shown in Fig. 3. The radiation of a train of USP, amplified with the aid of a ring amplifier^[6] to 2 J/cm^2 , was directed unto cell 3 with a layer of nitrobenzene 7 cm thick. Diaphragm 2 ahead of the cell, with diameter

FIG. 4. Dependence of the energy E falling in the aperture of the calorimeter on the energy density ϵ at the output of the cell: O-calorimeter at a distance 6 m from the cell with nitrobenzene, Δ -calorimeter at 0.1 m from the cell, \bullet -no cell with nitrobenzene. Dashed line-calculated dependence in the case when there is no cell with nitrobenzene.



10 mm, limited the beam cross section. The radiation energy passing through the cell was measured with calorimeter 4 having an imput aperture of 20 mm. The calorimeter was placed at a distance 6 m from the cell. In the absence of the cell, the entire energy of the laser beam entered the calorimeter. The energy density at the input of the cell could be decreased with the aid of calibrated filters 1.

The dependence of the energy E entering the aperture of the calorimeter on the energy density ϵ at the entrance to the cell is shown in Fig. 4. In the case when the calorimeter was located directly behind the cell, the registered energy equalled, within the limits of measurement accuracy, the energy at the entrance to the cell. This means that in the nitrobenzene the USP laser radiation is scattered through a rather large angle without an appreciable decrease of the total energy. This result enables us to state that the most probable cause of the attenuation of the laser beam in nitrobenzene is self-focusing, and not other nonlinear effects such as stimulated Raman scattering (SRS). Favoring this conclusion are also the data of^[9,10], where it is stated that the efficiency of conversion of laser radiation of ultrashort duration into SRS is quite low in the case of nitrobenzene, even under conditions of self-focusing of the laser beam.

Attention is called to the character of the curve of Fig. 4: with increasing density of the output flux of the USP train, the energy registered by the calorimeter first increases and then decreases. The decreasing part of the curve, in our opinion, indicates that in our experiment the loss-producing changes in the medium are subject to inertia.

Thus, the obtained data offer evidence that the occurrence of nonlinear losses in nitrobenzene through which USP radiation passes is connected with selffocusing.

The self-focusing of the USP radiation can occur also in the active neodymium-glass elements of either generators or amplifiers. Evidence of selffocusing is the presence of long thin filament-like faults in the glass, which were observed by us in^[1]. Self-focusing in the active medium of the generator gives rise to nonlinear loss, which, like the loss in the cell with saturable absorber, limit the generation energy and hinder the formation of a deeply-intensitymodulated sequence of USP.

It was of interest to ascertain the degree to which the loss connected with self-focusing influences the operation of a USP amplifier. To this end, investigations were made of the passage through the amplifier of a pulse separated from a train of USP from a master generator. These investigations were per-



FIG. 5. Microscopic faults on the end surface of a rod of neodymium glass.



FIG. 6. Distribution of the intensity in the laser beam of an amplifier (print of radiation on the photographic plate installed 4 m away from the output of the amplifier stage, 20 mm diameter \times 700 mm), at energy density $\epsilon = 2 \text{ J/cm}^2$ (a) and 0.1 J/cm² (b).

formed on an improved variant of the setup described $in^{[1]}$, in which we were able to obtain a sufficiently homogeneous distribution of the radiation over the beam cross section and high directivity of the radiation (divergence not worse than 10^{-4} rad).

At energy densities $>1 \text{ J/cm}^2$ we observed in the neodymium glass (both with and without microscopic admixtures of platinum) the formation of bundles of filament-like faults filling the entire cross section of the rod. The filaments were terminated on the surface of the glass by microscopic faults with dimensions of several times ten microns (Fig. 5). With increasing radiation flux, the number of filaments increased. Separate investigations have shown that the length of the filamentary fault can reach several centimeters and can exceed the length of the USP in the glass.

It was noted that the laser beam of the amplifier is accompanied at the exit from the last stages of the apparatus by a halo of scattered radiation (Fig. 6); the angular dimensions of the halo greatly exceeded (by 3-4 orders of magnitude) the divergence angle of the main beam. Special investigations have shown that the radiation of the halo, as well as the radiation of the main laser beam, is made up of pulses of ultrashort duration. The fraction of the laser energy dissipated in the halo greatly increased when the energy density was increased, and became comparable with the energy of the laser beam. The greatest intensity of the halo



FIG. 8. Time structure of USP radiation in amplification: a- oscillogram of input and output signals in the absence of nonlinear losses in the amplifier; b, c-oscillograms of input and output signals in the presence of nonlinear losses in the amplifier.

FIG. 9. Temporal structure of input and output signals registered by a photorecorder (case of nonlinear losses in the amplifier).

was observed when filamentary faults were produced in the glass. The effective gain K (the ratio of the laser-beam energy at the output to its energy at the input) then decreased sharply.

Figure 7 shows the dependence of K on the energy density of the input signal ϵ , taken for two amplifier stages with rods of LGS-228-2 glass of 20 mm diameter and 700 mm length. It is seen from the curve of Fig. 7 that when the energy density at the input is increased from 10^{-4} to 10^{-1} J/cm², the effective gain decreases by a factor of more than 100. The energy density at the output (~1 J/cm^2) was smaller by almost one order of magnitude that the saturation energy in the neodymium glass^[11]. This shows convincingly that the observed nonlinearity of the amplifier (Fig. 7) is not connected with the effect of saturation in the neodymium glass. As is evident from the experimental data, the nonlinearity is due to loss of radiation in the active medium of the amplifier, connected with selffocusing.

Simultaneous observation of the temporal structure of the input and the output signals with the photoelectronic recorder and an oscilloscope has shown that the time structure of the amplified signal changed strongly in the case of amplification accompanied by occurrence of non-linear losses. Additional pulses (Figs. 8 and 9) appear in the structure of the output signal. In the case of a weak signal at the input (less than 10^{-3} J/cm^2). no such change occurs (Fig. 8a). This result points to an important circumstance, namely, that the radiation of a USP neodymium-glass laser contains additional pulses of low intensity. Additional pulses of low intensity were observed earlier in the radiation of a ruby USP laser^[12]. Owing to the limited dynamic range of the apparatus, such additional pulses are difficult to observe. However, an amplifier with nonlinear losses

makes it possible to reveal these low-intensity pulses. Their presence in the laser radiation is one more fact evidencing the fluctuation mechanism of the formation of the $USP^{[2-4]}$.

We have thus established that in neodymium glass there exist losses that depend on the power of the transmitted USP, and that these losses are connected with the occurrence of self-focusing.

INFLUENCE OF NONLINEAR LOSSES ON THE OPERATION OF USP GENERATORS AND AMPLIFIERS

The results of the investigations show that selffocusing in an amplifying medium and the associated loss and damage to the medium are the factor that limits the growth of the density of the light flux in powerful USP amplifiers. Let us examine the influence of self-focusing and of losses connected with it on the operation of USP laser. The formation of the USP in a laser Q-switched by a nonlinear absorber with short relaxation time of the bleached state occurs, in accordance with^[13], in the following manner.

The generation begins with excitation of a large number of axial modes of the resonator with a random phase distribution. The interference of these modes leads to the appearance of intensity spikes of ultrashort duration. By successive passages of the radiation between the resonator mirrors, the distribution of the spikes becomes quasiperiodic, and the intensity of the spikes increases exponentially. Owing to the narrowing of the spectrum, the duration of the spikes also increases. This continues until the intensities of the spikes reach a level at which bleaching of the nonlinear absorber begins. Owing to the short relaxation time of the bleached state of the absorber, the most intense spikes become compressed and are amplified more rapidly than the remaining less intense ones. This gives rise to one or, in principle, several ultrashort pulses that are amplified during successive passages through the laser and ultimately radiate the energy stored in the active medium. As a result, there appears in the output of the generator a periodic sequence of intense USP, which are accompanied by a background of low intensity, also consisting of USP.

The presence of linear losses, either in the active medium or in other optical elements of the laser, can influence the picture of the development of the USP in the following manner. Starting with a certain intensity level, the most intense spikes cease to become amplified and are compressed more rapidly than the remaining ones. Thus, the nonlinear losses limit the process of separation of the most intense spikes. This leads to a decrease in the difference between the intensities of the main USP and the background USP. As a result, the probability of the appearance of a sequence of single USP decreases. In addition, a deterioration can occur in the temporal structure of the pulses in the USP train. Let us assume that at the start of the train there was produced an ultrashort pulse accompanied by weak satellites. Under conditions of nonlinear limitation, the intensities of these satellites increase relative to the main pulse at the end of the development of the train. This explains the results of^[5], where they

observed additional pulses on the axial period at the end of the train of USP generation. Owing to the finite relaxation time of the bleached state of the nonlinear absorber, the most favorable conditions for the increase of the intensity turned out to be those for the weak satellites that follow immediately the main USP. The time resolution of the apparatus (photocell and oscilloscope) may turn out to be insufficient to observe all these pulses. The oscilloscope screen will show a sequence of single pulses, each of which is proportional to the radiation energy within the limits of the time resolution of the apparatus. But owing to the deterioration of the time structure, the pulses having identical amplitudes on the oscilloscope screen at the beginning and end at the end of the train will have different peak powers, with the higher power in the pulses at the beginning of the train. This explains the results of investigations on the excitation of SRS and on the multiphoton photoeffect by USP radiation, where it was noted that the USP action is more effective at the beginning of the train.

The influence of the nonlinear losses on the operation of the laser will be the stronger, the larger the radiation intensity in the resonator. For this reason, attempts to increase the pulse energy in the USP laser by increasing the dimensions of the active medium and by using a denser nonlinear absorber should lead to a deterioration of the temporal structure of the radiation. This is confirmed, for example, by the results of $^{\hbox{\scriptsize [14]}}$, in which a neodymium-glass laser with mode locking was investigated, using active rods of length 16.5, 45, and 76 cm and a cell with a saturable dye of thickness 1 cm. When the 76-cm rod was used, the output energy in six pulses reached 44 J. The duration of each of these pulses reached 2 nsec. When the 45-cm rod was used, the duration was 0.75 nsec. Direct methods for determining the pulse duration, based on nonlinear optical phenomena, have shown^[14] that the obtained pulses consist of groups of closely-lying pulses of picosecond duration. When the 16.5-cm rod was used, the duration of the USP ranged from 2 to 10 psec.

Attempting to decrease the influence of the nonlinear losses on the operation of the USP generator, we decreased the energy density in the resonator to a value lower than $5 \times 10^{-3} \text{ J/cm}^2$. In this case, the probability of obtaining a sequence of single USP was not less than 90%, and the duration of the USP did not exceed 2×10^{-11} sec (the time resolution of the photorecorder).

CONCLUSION

We have observed and investigated nonlinear losses arising both in USP generators and USP amplifiers. We have shown that the cause of the nonlinear losses is the occurrence of self-focusing in the active medium as well as in the optical media of the other elements contained in the generators and amplifiers (cells with saturable dye, electrooptical shutters, prisms, plates, etc.). These losses exert a strong influence on the operation of the USP generators and amplifiers.

In the case of USP generators they lead to a deterioration of the temporal structure of the radiation, making it more difficult to obtain intense single USP. Since the nonlinear losses connected with self-focusing increase with increasing flux intensity and with increasing length of the medium, it is necessary, to improve the operation of the USP generator, to decrease the intensity of the flux inside the resonator and to decrease the lengths of the optical elements (active rods, cells).

The occurrence of self-focusing in rods of amplifiers is the cause limiting the growth of the USP energy. To increase the USP energy it is necessary to increase the cross sections of the output stages, and also to strive to obtain the necessary gain in as short an active-medium length as possible.

The presence of nonlinear losses in the amplifier has made it possible to reveal the complicated structure of the radiation of the USP generator. It turned out that intense pulses of the USP generator were accompanied by a background consisting of USP of lower intensity. An amplifier with nonlinear losses can thus be used to reveal the temporal structure of the generator. In particular, it can be used to determine the extent to which the intensity of the main USP exceeds the intensity of the background pulses.

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¹N. G. Basov, P. G. Kryukov, Yu. V. Senatskiĭ, and S. V. Chekalin, Zh. Eksp. Teor. Fiz. 57, 1175 (1969) [Sov. Phys.-JETP 30, 641 (1970)]. ²V. S. Letokhov, ibid. 55, 1077 and 1943 (1968) [28, 562 and 1026 (1968)].

³J. A. Fleck, Jr., Phys. Rev., B, 1, 84 (1970).

⁴ T. N. Kuznetsova, Zh. Eksp. Teor. Fiz. 57, 1673 (1969) [Sov. Phys.-JETP 30, 904 (1970)].

⁵ V. V. Korobkin, A. A. Malyutin, and M. Ya. Shchelev, ZhETF Pis. Red. 11, 168 (1970) [JETP Lett. 11, 103 (1970)].

⁶N. G. Basov, P. G. Kryukov, V. S. Letokhov, Yu. A. Matveets, and S. V. Chekalin, ibid. 10, 479 (1969) [10, 308 (1969)].

⁷ T. V. Volyak, S. D. Kaltmazov, A. A. Medvedev, and I. V. Pogorel'skii, Kratkie soobshcheniya po fizike (FIAN) (Brief Reports of Physics), No. 4, 15 (1970).

⁸M. E. Mack, R. L. Carman, J. Reintjes, and N. Bloembergen, Appl. Phys. Lett., 16, 209 (1970).

⁹R. G. Brewer and C. H. Lee, Phys. Rev. Lett., 21, 267 (1968).

¹⁰M. J. Colles, Optics communications, 1, 169 (1969).

¹¹N. G. Basov, V. S. Zuev, P. G. Kryukov, S. Letokhov,

Yu. V. Senat-skii, and S. V. Chekalin, Zh. Eksp. Teor.

Fiz. 54, 3 (1968) [Sov. Phys.-JETP 27, 1 (1968)].

¹²N. G. Basov, P. G. Kryukov, and V. S. Letokhov, Paper delivered at Conference on Nonlinear Optics in Belfast, September, 1969. FIAN Preprint No. 122, 1969.

¹³V. S. Letokhov, FIAN Preprint No. 109, 1969.

¹⁴ A. J. De Maria, W. H. Glenn, Jr., M. J. Brienza, and M. E. Mack, PIEEE, 57, 2 (1969).

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