Variation in the time structure of ultrashort pulses during propagation through a stable two-component medium

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It is shown that a stable two-component medium can be used to reduce the length of ultrashort pulses and the background intensity. It is established that the self-focusing of laser radiation in glass results in a substantial change in the time structure of ultrashort pulses and, in particular, each pulse is split into a number of components.

It has been established experimentally [1-4] that ultrashort pulses (USP) emitted by a neodymium glass laser have a complicated form and their total length substantially exceeds the reciprocal of the width of the radiation spectrum. Moreover, the main USP are accompanied by weaker satellites.^[5] This is connected with the finite relaxation time of the absorber through which radiation is passed, and with the nonlinear losses arising during the interaction between the radiation and the optical materials in the apparatus. These losses weaken predominantly the stronger USP^[6] and this leads to a reduction in the contrast of the radiation.

The effect of nonlinear losses decreases with decreasing field density in the laser cavity. This can be achieved, for example, by increasing the initial transmission of the nonlinear absorber. $[^{6}, ^{7}]$ However, there is then a substantial fall in the output energy of the USP generator, and the nonlinear losses remain the same as when an absorber with maximum possible initial transmission is employed. $[^{7}]$ Therefore, to increase the USP contrast (ratio of mean USP intensity to the background intensity), we use a USP amplification system consisting of a stable two-component medium. $[^{7-9}]$

The stable two-component medium (STM) consists of an amplifier and a nonlinear absorber with a short relaxation time of the transmitting state. The stability of the medium means that it is below the self-excitation threshold. A medium of this kind has well-defined discriminating properties, since it absorbs weak signals and amplifies strong signals. Moreover, the STM need not satisfy the self-excitation conditions, and this means that one can use a nonlinear absorber with a low initial transmission and obtain a high degree of discrimination in a single pass. The amplifying component should then compensate only the relatively small losses in the transmitting absorber. In the generator the reduction in the transmission of the absorber necessarily requires a corresponding increase in the gain and this, in its turn, leads to a reduction in the time of nonlinear interaction between the radiation and the absorber. It is well known that during this time there is an effective discrimination against fluctuation pulses, which leads to the formation of a chain of USP.^[10, 11] The nonlinear absorption time can be increased in the STM by ensuring that the loss is close to the gain.

In this paper, we investigate the interaction between the USP and the STM. The apparatus consists of the USP generator, an electrooptical shutter used to isolate a single USP from a chain, an STM based on a ring system, and a shutter for isolating a single pulse from the STM (Fig. 1). An isolated pulse from the generator is sent into the STM. To produce the maximum ratio of the main pulse power to the power of the background pulses (contrast), the input signal and initial transmission of the absorber in the STM are chosen so that the system operates with a slight excess over the threshold for the strongest pulses, i.e., for the main pulse the gain only slightly exceeds the losses. Successive passes through the STM produce either an increase or a reduction in the input signal, depending on its intensity.

Figure 2 shows oscillograms of the input and output signals. It illustrates the case when the input signal consists of two pulses of different amplitude. As can be seen, the output signal does not contain the second pulse with the lower amplitude. Estimates based on this oscillogram show that the amplitude ratio has increased by at least two orders of magnitude and the pulse energy is increased by a factor of 100 after 15 passes through the STM.

It is known that the STM should change the pulse shape.^[9,12,13] It is interesting to consider the change in the complicated time structure of the individual USP on passing through the STM. When the relaxation time of the nonlinear absorber is sufficiently small, one might hope to isolate one of the pulses in the complicated substructure of the USP from the laser with mode locking, and shorten it still further.

It is important to emphasize that the correct choice of the STM parameters is not a simple experimental



FIG. 1. Block diagram of the apparatus: 1 - active elements (neodymium glass), 2 - compensating lenses, 3 - aperture, 4 - glass wedge, 5 - solution of dye No. 3955 in nitrobenzene, 6 - Pockels shutter, 7 - spectrograph. Initial transmission of dye in generator -50%, in STM - 15%.



FIG. 2. Oscillogram showing input (arrowed) and output pulses in the STM.

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problem. The problem does not merely reduce to a choice of a state in which a chain of USP of equal amplitude is taken from the exit of the STM. Figure 3 shows the nonlinear characteristic of the transmitting absorber. The STM can be designed so that Pin is maintained equal to P_1 , P_2 , or P_3 . In all cases corresponding to the threshold state, one observes a sequence of equal USP's at the exit from the STM. However, in the case of P_1 , the STM will not exhibit any discrimination or shortening of the USP's since losses in the absorber are the same for all the pulses. For the ring STM which we have used, this state corresponds to the self-excitation threshold (the medium ceases to be stable). In the case of P_3 , there is again no shortening of the USP and the discrimination is weak since only low-intensity pulses are damped out by the absorber. This situation can be used to increase the USP contrast for sufficiently high contrast of the input signal. In the case of P_2 , both the shortening of the pulse and the discrimination are at their highest level.

Figure 4 illustrates a numerical calculation of the shortening of the USP, using formulas describing the action of a two-component medium.^[9] The parameters used in the computation are the initial transmission and the STM gain used in the experiment. The input signal is taken in the form of a single Gaussian pulse whose length is much greater than the relaxation time of the nonlinear absorber. It is clear from curve 1 that when the input pulse power is at the threshold level, it is shortened by a factor of 5 after ten passes through the STM. Departure from the threshold value is found to reduce this factor and to reduce the duration of the non-linear stage (curves 2 and 3). It is important to note that



FIG. 4. Calculated dependence of the relative reduction in the pulse length τ_N/τ_1 as a function of the number N of transits through the STM. Curve 1 corresponds to the input signal P = P_{th}, curve 2 to P = 0.5 P_{th}, and curve 3 to P = 2P_{th}. Curves 1', 2', and 3' were obtained for similar input pulses but with STM gain increased by 15% (τ_1 input pulse length, τ_n pulse length after N transits).

when $\mathbf{P} < \mathbf{P}_{th}$ the pulse shortening process is more effective during the first few passes as compared with the case $P = P_{th}$ (curve 2, Fig. 4). This suggests that the working point of our STM was chosen so that P_3 $> P_{th} > P_2$ (Fig. 3). When $P < P_{th}$, the pulse is damped out in the STM and falls into the range of intensities for which the shortening is faster (see Fig. 3). It then passes through the value P_2 and finally falls to P_1 . The nonlinear stage terminates at this point and there is no further shortening of the USP. When $P>P_{th},$ the non-linear stages are still faster (curve 3, Fig. 4). Figure 4 also shows the curves for different input signals with the STM gain increased by 15% (curves 1', 2', 3'). In practice, the STM always exhibits some gain, i.e., the input signal is slightly above the threshold. From the standpoint of the best discrimination and maximum shortening of the USP, it is most convenient to choose the STM parameters so that P_{th} lies to the left of P_2 (Fig. 3). In the course of the amplification process, the pulse will lie in the region of the nonlinear interaction with the absorber for a longer period of time. However, Pth must be greater than P_1 so that weaker signals and intrinsic STM noise are not amplified.

To investigate the change in structure and duration of the USP's after passing through the STM, we recorded the spectrum of the input and output signals. Figure 5 shows the results. The width of the spectrum of the USP chain is 5.4 cm^{-1} (Fig. 5a). When the USP is passed through the STM, the spectrum is much broader and smoother. The spectrum broadening is very dependent on the extent to which the amplification state approaches the threshold (for which the gain for the input-signal maximum is equal to the losses). Near the threshold the spectrum width is a maximum and reaches 34 cm^{-1} . In this case, there is also the greatest asymmetry of the envelope of the spectrum (Figs. 5b-e).

The spectrum may be broadened both as a result of the reduction in the duration and discrimination against the USP structure, and also due to effects associated with the nonlinearity of the refractive index. In the last case, the width of the spectrum increases with increasing intensity, and the spectrum itself becomes choppedup.^[14] In our case, the maximum broadening is observed near the threshold, and when the intensity is increased the width of the spectrum is found to fall (Fig. 5). The broadening of the spectrum must therefore be associated with the first two factors. The origin of the observed asymmetry of the envelope of the output spectrum is probably connected with the fact that the



FIG. 5. Emission spectra at input (a) and output (b-e) of the STM under different conditions: a – spectrum of the USP generator, $\Delta \nu = 5.4$ cm⁻¹; b – threshold conditions, $\Delta \nu = 34$ cm⁻¹, c – gain 1.5% above threshold, $\Delta \nu = 30$ cm⁻¹, d – gain 10% above threshold, $\Delta \nu = 26$ cm⁻¹, e – gain 15% above threshold, $\Delta \nu = 20$ cm⁻¹.

luminescence line of the neodymium glass does not coincide with the center of the absorption line of the dye. The maximum asymmetry in the threshold state must then indicate that the dye is most effective in this case.

The discriminating effect of the transmitting absorber is opposite to the effect of nonlinear losses. However, in the USP generator, the stage of nonlinear interaction between radiation and the absorber usually ends at a low-intensity level (~ 10 MW/cm²) for the solution of dye No. 3955 in nitrobenzene.^[15] The USP which carry the main fraction of energy of the chain completely saturate the absorber and are therefore practically not discriminated against. In this region, their characteristics deteriorate because of nonlinear losses. In the STM with correctly chosen parameters, the effect of the absorber competes with the nonlinear losses despite the high field densities (as compared with the generator).

We were not able to observe directly with sufficient time resolution the change in the structure and duration of the USP in the STM. Our studies were therefore carried out by an indirect method, namely, by determination of the change in the spectrum and the coefficient representing the transformation of the radiation into the second harmonic.^[7] The transformation into the second harmonic was carried out with a KDP crystal, 5 mm thick. We measured the transformation coefficient $K = I_{2\omega}/(I_{\omega})^2$ where I_{ω} is the USP energy at the fundamental frequency and $I_{2\omega}$ is the energy at the second-harmonic frequency. It is shown in^[7] that the coefficient K increases with decreasing number of subpulses in the USP substructure and with decreasing length of these pulses. The experimental dependence of K on the number of passes of the USP through the STM is shown in Fig. 6. It is clear that, in both cases, K increases with increasing number of passes through the STM; in case b which differs from case a by the fact that the input signal power is higher by an order of magnitude, the coefficient K shows certain discontinuities which are probably connected with the increased effect of nonlinear losses. When the transmitting dye is replaced in the STM by pure nitrobenzene, the factor K is found to fall on the average by a factor of three per pass.

If we compare the variation in K (Fig. 6) with the calculated reduction in the pulse length in the STM in the optimal case (Fig. 4, curve 1), we may conclude that the measured increase in K exceeds even the prediction for the maximum reduction in the length of the input signal. In all probability, the increase in K is connected not only with a reduction in the pulse length but also with the iso-



FIG. 6. Measured transformation coefficient K as a function of the number N of USP transits in the STM. The output power in case b is higher by an order of magnitude than in case a.

lation of the strongest components in the USP substructure. The fact that this process should proceed more effectively in the two-component medium than the reduction in length is shown theoretically in papers on the USP generator.^[10,11]

We associate sudden changes in K (Fig. 6b) with the change in the USP shape due to the interaction between the high-power radiation and the active medium (glass matrix). As the USP power increases, the refractive index of the glass acquires a nonlinear increment Δn which leads to the self-focusing of the radiation. The mechanism responsible for Δn in glass has practically zero inertia^[14] and, therefore, only the pulse maximum is at first self-focused (in time). The self-focusing leads to scattering of the radiation into a broad angle, thus taking it out of the aperture of the resonator. The scattering of the strongest part of the USP results in the appearance of a valley in the pulse. This process is repeated in the course of the successive USP passes through the active medium, the final result of which is that the pulse is split up into still shorter fragments.

We have carried out a numerical calculation on this process for the ring amplifier used in our experiments. The calculation has been carried out subject to very rough assumptions, namely, the effect of the nonlinear absorber, the dispersion in the active medium, the finite amplification bandwidth, and the diffraction effect at the aperture were all ignored. The active medium was replaced by a thin lens with a focal length of the form

$f_{\rm nonlin} = a^2/2n_2 |E_{\mu}(t)|^2 l \varkappa^2,$

where a is the input-beam diameter, n_2 is the nonlinear refractive index of glass (n₂ = 3×10^{-13} cgs esu), E_µ(t) is the maximum wave field within a given cross section at a given time, l is the length of the active medium, and κ is the gain in the active medium. It was assumed that losses during self-focusing occur as a result of the restriction of the light beam by the 7-mm diameter aperture. We calculated the change in the USP energy in successive transits through the STM (the form of the chain on the oscilloscope screen). We also calculated the change in K and in the shape of the USP itself. Some of the results of this calculation are shown in Fig. 7. The change in the USP due to self-focusing, the deformation of the pulse-chain envelope, and the change in K are all clear from the figure. In spite of the relatively rough assumptions used in the calculation, the nature of the change in the chain envelope and the change in K correspond to the experimental data: peaks on the envelope correspond to minima of K and maxima on the K curve correspond to minima on the envelope. The calculations do not predict a substantial increase in K since the effect of the nonlinear absorber is not taken into account. This can be done in the case of the USP generator but experiment shows that K usually falls off down the chain. This fall-off is associated with the presence of smaller satellites near the main USP and with the fact that these can be observed in the presence of nonlinear losses.^[6] These low-intensity satellites are not taken into account in the calculation, and this again leads to a discrepancy as compared with experimental data.

When the above mechanism for nonlinear losses in the STM is operative, the USP chain envelope should exhibit valleys (see Fig. 7) associated with the fact that a substantial fraction of the USP energy is scattered into a broad angle as a result of self-focusing. We used the

A. N. Zherikhin et al.

coaxial FÉK-15 photodiode to observe simultaneously the USP chain from the STM and the radiation scattered by the active medium in the forward direction for angles between 2.5 and 8° from the optical axis. The oscillograms obtained in this way (Fig. 8a) give a relatively good correlation between the minima on the main radiation and the maxima on the scattered radiation. Estimates based on these oscillograms show that up to 10%of the main radiation is scattered into this angular interval. Scattering into a cone with an apex angle of 2.5° is stronger by an order of magnitude, probably because of the amplification of the scattered radiation in the active medium. Hence, it may be concluded that self-focusing can be responsible for the scattering of a substantial or even predominant fraction of the pulse energy. Scattering in the backward direction has similar characteristic features but an intensity lower by a factor of 20. It is important to note that when the forward-scattered



FIG. 7. Calculated change in the USP energy E, transformation coefficient K, and USP shape (a-e) as functions of the number N of passes through the STM; a - USP shape after fifth pass, b - after eighth pass, c - after eleventh pass, d - after twenty-first pass.



FIG. 8. Variation in the main (a) and scattered (b) radiation in the STM. In case a the scattered radiation was observed through neutral light filters and in case b through SZS-14 and SZS-16 light filters.

intensity is observed through SZS-14 and SZS-16 filters, which reduce the radiation at 1.06 μ by a factor of almost 10⁷, one can also see the scattered USP's (Fig. 8b). In this case, they have the form of individual pulses at greater intervals than under the previous conditions (see Fig. 8a). For comparison, we note that when the oscillograms were recorded, the scattered radiation was received through neutral light filters reducing the 1.06 μ radiation by a factor of 5. This fact shows that the scattered radiation has a broad spectrum and its width changes on successive transits through the STM. This broadening of the spectrum is typical of self-focusing.^[17]

Experiments with the STM have thus shown that it can be used to shorten the length of ultrashort pulses and to increase their contrast. The shortening can occur in two ways. The input signal can be a smooth single pulse, but the rate of reduction in length of this pulse in the STM is relatively small (see Fig. 4). A more effective procedure is to use an input pulse split into short components as a result of nonlinear losses connected with self-focusing. The discriminating effect of the STM can, in principle, be then used to isolate one such component and thus obtain a single USP. It is important to note that, in both cases, the pulse length is restricted by the amplification bandwidth of the active medium.

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