GENERATION OF ULTRASHORT LIGHT PULSES IN A LASER WITH A NONLINEAR ABSORBER

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The formation and evolution of ultrashort light pulses in a laser with a saturable absorber is considered theoretically. The probability of a random intensity burst occurring at the expense of radiation-intensity fluctuations in the set of axial modes is estimated. It is shown that spreading of the initial fluctuation-intensity burst occurs in the linear generation region at the expense of the amplifying medium, whereas in the nonlinear region the pulse is compressed by the nonlinear absorber. In a laser with an infinitely rapid relaxation of the saturated state, the minimal duration of the pulse is several times larger than the limiting value and natural mode selection in the laser must be eliminated in order to attain the limiting value. The evolution of the pulse shape is considered in the case of an absorber with a finite relaxation time of the saturated state. It is shown that very rapid relaxation of the saturated state (10^{-13} sec) can occur at the expense of molecular transitions between excited singlet levels.

INTRODUCTION

MUCH progress was made recently in the field of generation of powerful ultrashort pulses of light by the method of self-phasing of axial modes of a solid-state laser saturated by an absorber[1,2]. A nonlinear absorber (usually a saturable solution of dye) Q-switches the laser and successively compresses the pulse propagating inside the cavity. This can be attained simultaneously if the lifetime T_{1b} of the excited absorber particles is much shorter than the time T necessary for the pulse to traverse the resonator ($T = 2L/v$, where L is the resonator length and v is the average group velocity of the light in the resonator), i.e., if the nonlinear absorber is "fast" or has low inertia. Self-compression of the pulse in the resonator is equivalent to a self-increase of the number of phased modes, which is usually called self-phasing or self-mode-locking. The efficiency of this method can be assessed by the fact that a neodymium-glass laser with a "fast" saturable solution generates a train of ultrashort pulses of duration on the order of 10^{-11}–10^{-12} sec and of energy on the order of 10^{-3} J[3,4].

A number of problems dealing with steady-state generation of a laser with a saturable absorber are considered in[5,6]. These investigations dealt with effects of the position of the absorber inside the resonator[5,6] and with the limitation of the pulse duration as a result of the finite lifetime T_{1b}[6]. Several questions in the development of ultrashort pulses in the laser with infinitely "fast" absorber ($T_{1b} = 0$) were considered in[7,8]. However, there is still no theoretical analysis capable of explaining the results of the existing experiments and of suggesting ways of reaching the limiting pulse duration, which is determined by the width of the amplification band. The purpose of the present paper is to fill this gap.

In this article we consider the occurrence and evolution of an ultrashort pulse in a laser with a nonlinear absorber, including all the stages of generation, viz., formation of the initial ultrashort intensity burst due to the fluctuations of the radiation intensity, broadening of the fluctuation burst in the linear region as a result of the finite amplification band width, and compression of the pulse in the nonlinear region as the result of the absorber. In particular, it is shown that to attain the limiting pulse duration it is necessarv to eliminate the broadening of the pulse in the linear region of development, using for example frequency-selective losses in the resonator. We consider the influence of the finite relaxation time T_{1b} of the saturated state of the absorber on the evolution of the pulse and mechanisms of obtaining extremely small values of T_{1b} for molecules in a solution. The proposed theory explains the results of the existing experiments and makes it possible to point to ways of obtaining powerful ultrashort pulses with duration on the order of 10^{-13} sec.

1. QUALITATIVE ANALYSIS

We consider the following model of a laser with "fast" saturable absorber. The resonator contains an excited medium with gain in a frequency band $\Delta\omega_a$ centered at the frequency ω_0, and an unexcited medium with absorption in a frequency band $\Delta\omega_b$, within which the amplification band is located. We shall henceforth denote the parameters of the amplifying medium by the index a, and that of the absorbing medium by b.

When the active medium is pumped, the gain compensates for the loss in the absorbing medium and in the mirrors at a certain instant of time ($t = t_0$). From that instant of time on the gain exceeds the loss, and the radiation intensity in the resonator modes increases in accordance with

$$\exp\left\{\frac{1}{T}\int_{t_0}^{t}[\alpha(t')-\alpha_0]\,dt'\right\},$$

where $\alpha(t)$ is the gain per pass of the active medium and $\alpha_0 = \alpha(t_0)$ is the threshold gain per pass. After a certain time τ_1 the intensity of the radiation becomes sufficient to saturate the absorption. The delay τ_1 of the instant when saturation occurs relative to the instant of the threshold is given by

$$\tau_1 = T\left[\frac{2\tau_p}{\alpha_0 T}\ln\left(\frac{I_s}{I_0}\frac{1}{\varkappa_0}\sqrt{\frac{\alpha_0 T}{2\tau_p}}\right)\right]^{1/2}, \tag{1}$$

where T is the time necessary for the light to travel through the resonator, τ_p is the time characterizing the growth rate of the gain as the result of the pumping when the threshold is exceeded ($\tau_p^{-1} = \alpha^{-1} d\alpha/dt|_{t=t_0}$), I_0 is the initial intensity determined by the spontaneous radiation in the modes, I_s is the intensity of absorption saturation, and κ_0 is the initial loss per pass in the absorber. For a two-level system with a particle lifetime T_{1b} at the upper level and a radiative transition cross section σ_b, the saturation intensity is determined by the expression

$$I_s = \hbar\omega_0 / 2\sigma_b T_{1b}. \tag{2}$$

At the instant of time $t \approx t_0 + \tau_1$ the loss in the absorbing medium decreases strongly—the Q of the cavity is self-switched[9-11]. This is followed by a faster increase of the intensity, and within a short time $\tau_2 \ll \tau_1$ by saturation in the amplifying medium ($\sigma_a < \sigma_b$), accompanied by emission of the stored energy in this medium in the form of a giant pulse.

Several axial modes are inevitably excited in the resonator,[1)] and the beats between them give rise to intensity fluctuations with a characteristic time $\sim T/m$, where m is the number of excited modes. The number of fluctuation bursts in the time interval T is of the order of m, and their amplitude is random. If the relaxation time T_{1b} of the saturated state of the absorber is much shorter than T/m, then the absorber "builds up" intensity fluctuations in the nonlinearity region. This is due to the fact that the losses in the absorber decreases with increasing instantaneous radiation intensity. As a result, the most intense fluctuation bursts become stronger and are compressed much more rapidly than the remaining ones, and ultimately they cause saturation of the absorption, and then amplification by the media in the resonator. Upon each reflection from the semitransparent mirror, part of the radiation leaves the resonator, and therefore the output radiation of the laser represents a sequence of ultrashort pulses with intervals T between them.

2. METHOD OF SOLUTION

A train of equidistant pulses of light obviously can be described by a set of axial modes with a definite phase relation between them. Such an approach, in principle, is equivalent to the approach as described above, but an analysis of the interaction of a very large number of modes is quite complicated and can be developed consistently only for the case of external periodic modulation of the losses (in the stationary[13] and nonstationary[14] generation regimes). In our case, a more suitable approach is the one based on the analysis of the change of the shape of the pulse and used for this purpose in a number of investigations[6-8,15].

Accordingly, we shall consider the propagation of a pulse in a resonator containing amplifying and absorbing media. The interaction between the pulse and the amplifying medium will be described in this case by the usual rate equations. Since the energy of the pulses is smaller by two or three orders of magnitude than the gain saturation energy $\mathscr{E}_{as}$, this means that we confine ourselves to the case when the pulse duration is $\tau \gg 2\pi/\Delta\omega_s$. We shall investigate the evolution of the pulse in this region and obtain the conditions under which the pulse duration is visible. If these conditions are satisfied, then the pulse duration is already limited by the finite gain bandwidth.

The interaction of the pulse with the absorbing medium will also be described by rate equations, assuming that $\Delta\omega_b \gg \Delta\omega_a$. Then the equations describing the transformation of the pulse per pass through the resonator are

$$\partial I_k(t) / \partial k = I_k(t)[\alpha_k(t) - \varkappa_k(t) - \gamma], \tag{3a}$$
$$\partial N_{ak}(t) / \partial t + (N_{ak} - N_a^0) / T_{1a} = -2\sigma_a I_k(t) N_{ak}(t), \tag{3b}$$
$$\partial N_{bk}(t) / \partial t + (N_{bk} - N_b^0) / T_{1b} = -2\sigma_b I_k(t) N_{bk}(t), \tag{3c}$$

where $-\frac{1}{2}T \le t \le \frac{1}{2}T$; $I_k(t)$ is the form of the pulse after the k-th pass; $\alpha_k(t)$ is the gain in the active medium in the k-th pass, and is connected with the density of the inverted level population $N_{ak}(t)$ by the relation $\alpha_k = \sigma_a N_{ak} L_a$ (L_a—length of the amplifying medium); $\kappa_k(t)$ is the loss in the absorbing medium in the k-th pass, connected with the density of the absorber level population difference $N_{bk}(t)$ by an analogous relation, $\kappa_k = \sigma_b N_{bk} L_b$ (L_b—length of absorbing medium); γ is the linear radiation loss per pass, and N_a^0 and N_b^0 are the stationary level-population differences in the amplifying and absorbing media at zero field amplitude. The populations N_{ak} and N_{bk} at the start of the k-th pass are obviously equal to the populations after the (k − 1) st pass:

$$N_{ak}(t - T/2) = N_{a,k-1}(t + T/2), \quad N_{bk}(t - T/2) = N_{b,k-1}(t + T/2).$$

Equation (3a) is valid in the approximation in which the change of the pulse shape per pass is small. Since the energy of the pulse is not sufficient to saturate the gain in one pass ($\mathscr{E} \ll \mathscr{E}_{as}$), this condition imposes a limitation only on the magnitude of the saturable loss:

$$\varkappa_0 \ll 1. \tag{4}$$

Condition (4) is satisfied with sufficient accuracy in lasers of ultrashort pulses, for which usually $\kappa_0 \approx 30\%$[1,2].

The evolution of the pulse is best considered in two successive stages: occurrence and evolution of the intensity fluctuation bursts in the region of the linear absorption, and the deformation of the pulse in the region of nonlinearity of the absorption. In turn, the evolution of the pulse in the nonlinear region is best considered in two stages that follow each other:

$$\tau \gg T_{1b}, \tag{5a}$$
$$\tau \approx T_{1b}. \tag{5b}$$

1) An exception is the case when frequency-selective elements are present in the resonator. Their influence on the number of excited axial modes was investigated in [12].

3. FORMATION AND EVOLUTION OF PULSE IN THE LINEAR REGION

Let the spectrum of the initially excited axial modes have a width $\Delta\omega_0$. The emission phases in the modes are random, and consequently the instantaneous emission intensity in the modes $I(t)$ is a random function of the time. The average duration of the intensity fluctuation is $\tau_f \approx 2\pi\Delta\omega_0^{-1}$. If the number of the excited modes is $m \gg 1$, then the total emission can be assumed to be similar to incoherent radiation[16]. The distribution function of the intensity fluctuations is given by the relation[16,17]

$$W(I)=\frac{1}{\langle I\rangle}\exp\left(-\frac{I}{\langle I\rangle}\right). \tag{6}$$

The probability of an intensity fluctuation with an amplitude exceeding the mean value $\langle I\rangle$ by a factor β is given by the relation

$$\mathcal{P}(\beta)=\int\limits_{\beta\langle I\rangle}^{\infty} W(I)\,dI=e^{-\beta}. \tag{7}$$

The frequency of the appearance of the intensity bursts is $\sim 1/\tau_f$. Consequently a fluctuation burst with amplitude $\beta\langle I\rangle$ occurs on the average, with unity probability, within a time interval $\tau(\beta)$:

$$\tau(\beta)\approx\tau_f e^{\beta}=\frac{2\pi}{\Delta\omega_0}e^{\beta}. \tag{8}$$

A fluctuation bursts with an amplitude 10 times the mean value will appear, for example, in a neodymium laser at $\Delta\omega_0 = \Delta\omega_a$[5] within an average time $\tau(10) \approx 10^{-8}$ sec. On passing through the threshold, amplification of the pulse sets in. The pulse is amplified by e times within a time τ_{amp} after passing through the threshold; this time is given by the expression

$$\tau_{amp}\approx(\tau_p T/2\alpha_0)^{1/2}. \tag{9}$$

The condition for the occurrence of a distinct intensity fluctuation burst, which can be the initial pulse in the laser, can be written in the form

$$\tau(10)\lesssim\tau_{amp} \tag{10}$$

In a laser Q-switched by a saturable absorber, satisfaction of this condition is quite feasible[2)].

In the region of linear development, natural mode selection by the amplifying medium takes place, owing to the preferred amplification of the modes near the center of the gain line. Natural selection is particularly important in lasers with saturable absorber, owing to the large delay time τ_1[19]. The selection causes the spectrum of the excited modes to become narrower during the delay time or, in other words, the initial intensity fluctuations broaden. This must be kept in mind when estimating the duration of the ultrashort pulses[20]. If τ_0 is the duration of the initial fluctuation intensity burst, then the time τ_0' to the instant of saturation of the absorption becomes equal to

$$\tau'=\tau_0\left[1+\alpha_0\frac{\tau_1}{T}\left(\tau_0\frac{\Delta\omega_a}{8}\right)^{-2}\right]^{1/2}, \tag{11}$$

where, for simplicity, the shape of the initial fluctuation and the shape of the gain line as assumed to be Gaussian. The quantity $8/\Delta\omega_a$ equals the limiting pulse duration τ_{lim}, determined only by the width of the amplification band. If $\tau_0 \ll \tau_{lim}\sqrt{\alpha_0\tau_1/T}$, then the fluctuation duration at the end of the linear development does not depend on the initial duration:

$$\tau'=\tau_{lim}\sqrt{\alpha_0\tau_1/T}. \tag{12}$$

Expression (12) determines the minimum duration of the fluctuation burst at the instant of the start of this compression by the nonlinear absorber.

4. EVOLUTION OF THE PULSE IN THE REGION $\tau \gg T_{1b}$

If the pulse duration $\tau \gg T_{1b}$, then the transparency of the absorber is determined by the instantaneous value of the intensity $I(t)$. In addition, in this case we certainly have $T \gg T_{1b}$, and consequently the absorber has time to relax to the initial state, with absorption κ_0, before the next passage of the pulse. During the first stage, obviously, it is possible to neglect the gain saturation. Then the change in the form of the pulse is described by the single equation

$$\frac{\partial P_k(t)}{\partial k}=P_k(t)\left[\alpha_k-\frac{\varkappa_0}{1+P_k(t)}-\gamma\right], \tag{13}$$

where we have introduced the dimensionless intensity $P = I/I_s$, I_s being the saturation intensity defined in (2).

The dependence of the gain on k is due to the pumping effect during the delay time τ_1. This change is quite small and can be represented in the form

$$\alpha(k)=\alpha_0+\left.\frac{d\alpha}{dt}\right|_{t=t_0}Tk, \tag{14}$$

where $\alpha_0 = \alpha(t_0) = \gamma + \kappa_0$ is the threshold gain. The most significant circumstance in (14) is that $\alpha_k > \alpha_0$, i.e., the gain exceeds the loss. Rather than consider the change of $\alpha(k)$, it is sufficient to take into account the main consequence of this change—the difference between $\alpha(k)$ and the threshold value α_0. To this end, it is natural to introduce the value of $\alpha(k)$ averaged over the delay time τ_1:

$$\alpha=\alpha_0+\frac{d\alpha}{dt}\frac{\tau_1}{2}=\alpha_0\left(1+\frac{\tau_1}{2\tau_p}\right), \tag{15}$$

where τ_p is the constant introduced above, which characterizes the pumping rate at the instant of the threshold; τ_1 is the delay time defined in (1). At a constant value of α, Eq. (13) can be integrated and its solution $P_k(t)$ is determined implicitly by the expression

$$\begin{aligned}&P_k^{-1}(t)\left[(\alpha-\varkappa_0-\gamma)+\alpha P_k(t)\right]^{(\varkappa_0+\gamma)/\alpha}\\&=P_0^{-1}(t)\left[(\alpha-\varkappa_0-\gamma)+\alpha P_0(t)\right]^{(\varkappa_0+\gamma)/\alpha}e^{-(\alpha-\varkappa_0-\gamma)k},\end{aligned} \tag{16}$$

where $P_0(t)$ is the initial state of the pulse.

Let us determine the effective duration of the pulse τ_k by the relation

$$\frac{1}{\tau_k^2}=-\frac{1}{P_k(t)}\left.\frac{\partial^2 P_k(t)}{\partial t^2}\right|_{t=0}, \tag{17}$$

where $t = 0$ is the point of the maximum of the pulse, which is constant in this approximation. Differentiating (16) twice at the point $t = 0$, we obtain after some

[2)]The considered mechanism of formation of fluctuation bursts also explains fully the spontaneous formation of picosecond pulses in solid-states without a nonlinear absorber, observed in [18].

transformations

$$\tau_k^2 = \tau_0^2 \frac{1+P_k}{1+P_0} \frac{(\alpha-\varkappa_0-\gamma)+\alpha P_0}{(\alpha-\varkappa_0-\gamma)+\alpha P_k}, \tag{18}$$

where P_k and T_0 are the intensities at the maximum ($t = 0$). Since the intensity of the pulse $P_0(t = 0)$ certainly satisfies the condition $P_0 \ll (\alpha - \kappa_0 - \gamma)/\alpha$, the change of the duration is given by

$$\tau_k^2 = \tau_0^2(1+P_k)\left(1+\frac{\alpha}{\alpha-\varkappa_0-\gamma}P_k\right)^{-1}. \tag{19}$$

From (19) it follows that the compression of the pulse is finite, even at an infinitely rapid relaxation of the absorber. Indeed, putting $P_k \gg 1$, we obtain the minimum pulse duration:

$$\tau_{min} = \tau_0\left(\frac{\alpha-\varkappa_0-\gamma}{\alpha}\right)^{1/2} = \tau_0\left(\frac{\tau_1}{2\tau_p}\right)^{1/2}. \tag{20}$$

Finally, the maximum pulse compression is determined by the following expression:

$$\frac{\tau_{min}}{\tau_0} = \left[\frac{T}{2\alpha_0\tau_p}\ln\left(\frac{I_s}{I_0}\frac{1}{\varkappa_0}\sqrt{\frac{\alpha_0 T}{2\tau_p}}\right)\right]^{1/2}, \tag{21}$$

where we have used expression (2) for the delay time τ_1. It follows from (21) that the compression increases with increasing τ_p, i.e., in the case of a slower passage through the threshold. The maximum possible value of τ_p is limited by the technical requirement that the delay time τ_1 be noticeably smaller than the duration of the pumping and the amplification relaxation time T_{1a}.

Let us consider the following numerical example: $\tau_1 \approx 3\times10^{-4}$ sec, $T \approx 6\times10^{-9}$ sec, $\alpha_0 \approx 0.5$, $\kappa_0 \approx 0.2$, and $I_0/I_s \approx 10^{-10}$. In this case $\tau_p = 0.25$ sec and τ $\tau_{min}/\tau_0 \approx 2.5\times10^{-2}$. It is clear therefore that to obtain pulses with duration shorter than 10^{-12} sec it is necessary to have $\tau_0 < 0.5\times10^{-10}$ sec, i.e., it is necessary that more than 10^2 axial modes take part in the formation of the fluctuation peaks at the start of the nonlinear development. However, their number is limited by the natural mode selection. If we choose as τ_0 the duration of the fluctuation peak at the end of the linear development (12), then the minimum duration of the pulse at the end of the contraction, according to (21), is determined by the relation

$$\tau_{min} = \tau_{lim}\left[\ln\left(\frac{I_s}{I_0}\frac{1}{\varkappa_0}\sqrt{\frac{\alpha_0 T}{2\tau_p}}\right)\right]^{1/2}. \tag{22}$$

At standard laser parameters $\tau_{min} \approx (4-5)\tau_{lim}$. Thus, in the ideal case of complete absence of selective elements in the resonator and an infinitely "fast" linear absorber, the minimum duration of the pulse is 4—5 times larger than the limiting value determined by the width of the amplification line.

5. GENERATION OF PULSES OF LIMITING DURATION

In order to attain the limiting pulse duration τ_{lim} it is necessary to suppress the natural selection of the axial modes of the amplifying medium. This can be done by eliminating the maximum gain at the center of the line, for example, by the following method. Let us introduce into the resonator a linear absorbing medium, whose absorption line center coincides with the center of the amplification line ω_0, and whose absorption-band width is somewhat smaller than the width of the amplification line $\Delta\omega_a$. If the additional absorption is less than α_0, then the summary line of the effective amplification of these two media has a minimum at the center. It is therefore obvious that the mode spectrum broadens rather than narrows. The duration of the intensity fluctuation burst in a laser with such an additional resonant exciter should not increase during the linear development of the generation, i.e., τ' can be of the order of τ_0 and the minimum pulse duration may equal τ_{lim}.

We note that in the proposed ultrashort-pulse laser the nonlinear absorber no longer plays the role of an element that compresses the light pulse. The nonlinear absorber only separates the most intense fluctuation burst against the background of the remaining less intense bursts. Naturally, to this end it is necessary to have $T_{1b} \lesssim \tau_{lim}$. If $T > \tau_{lim}$, then the fluctuation bursts with smaller amplitude, which follow the most intense one in the interval $\sim T_{1b}$, will also be amplified in addition to the intense fluctuation burst of limiting duration. As the result, the ultrashort pulse with duration $\tau \ll T_{1b}$ may have a weak but prolonged tail.

If the pulse duration becomes comparable with the relaxation time of the saturated state of the absorber T_{1b}, it is necessary to take into account the inertia of the absorber.

6. EVOLUTION OF THE PULSE IN THE REGION $\tau \sim T_{1b}$.

The evolution of the pulse, including the region in which the inertia of the absorber becomes appreciable, can be determined by solving exactly the system of equations (3). We note that pulse propagation in a nonlinear absorbing medium with relaxation of the saturated state was investigated by Zuev and Shcheglov[21] and later discussed in a paper by Khartsiev et al.[22] Our case differed in that the gain greatly exceeds the loss, at least at those instants when $\tau \sim T_{1b}$.

The main effect of the finite relaxation time of the saturated state of the absorber consists in the fact that the absorber does not have time to "collapse" on the trailing edge of a pulse of duration $\tau < T_{1b}$. As the result, absorption occurs only on the leading front, and the trailing edge of the pulse remains practically undeformed. Therefore the shape of a pulse of duration $\lesssim T_{1b}$ becomes asymmetrical.

We present first the results of a computer integration of the complete system of equations (3). In these calculations we took into account simultaneously the effect of the gain saturation. Such a model is already close to a real laser. We present the solution for the case when the parameters of the laser are such that the pulse duration τ_{min} with an infinitely "fast" absorber is several times shorter than T_{1b}. In this case the pulse duration is limited precisely by the delay time of the absorber T_{1b}. Figure 1a shows the change of the duration τ and of the peak power P_m of the pulse, and also the change of the gain of the active medium, as functions of the number of passes through the resonator k, and Fig. 1b shows the shape of the pulse for three characteristic instants of generation development: start, maximum, and end of giant pulse.

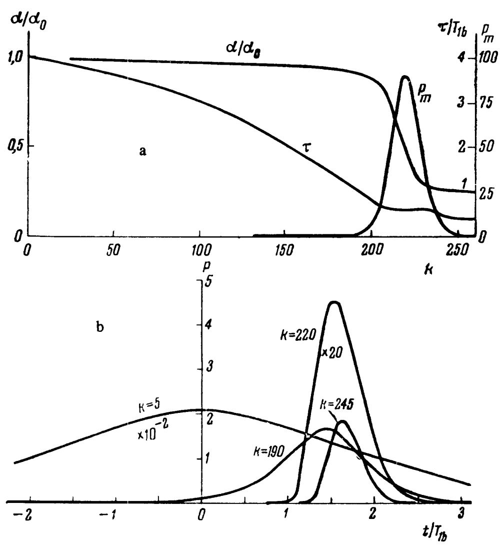

FIG. 1. Development of generation of ultrashort pulse: a—pulse duration τ, maximum power P_m, and gain α of the active medium as functions of the number of passes through the resonator k (the number of passes is counted arbitrarily); b—shape of ultrashort pulse at different instants of generation (k = 5, 190, 220, 245). Laser parameters: $\sigma_a/\sigma_b = 10^{-3}$; $\kappa_0 = \gamma = 0.3$; $(\alpha-\alpha_0)/\alpha_0 \approx 3 \times 10^{-2}$ at k = 0. The peak pulse intensity is $P_m \approx 2 \times 10^{-2}$ at k = 0.

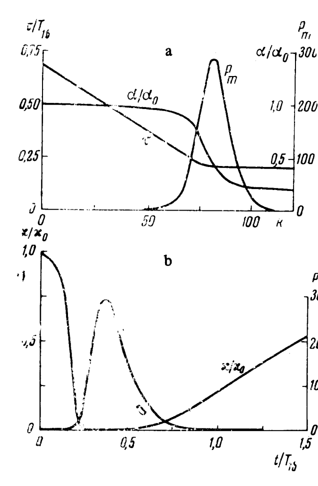

FIG. 2

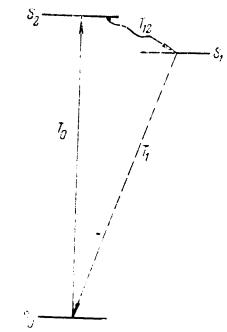

FIG. 3

FIG. 2. Development of generation of ultrashort pulse in the case when the pulse duration is limited by the inertia of the absorber: a—change of duration and of the peak power of the pulse and of the gain of the medium as a function of the number of passes k ($\sigma_a/\sigma_b = 10^{-3}$); b—shape of the pulse and change of absorption under the influence of this pulse at k corresponding to the maximum peak power of the pulse.

FIG. 3. Diagram of two-step relaxation of the saturated state of the dye solution.

With this as an example, we can see clearly the main features of the evolution of the pulse, which were considered above and in our earlier paper[8]. First, it is seen in Fig. 1 that the compression of the pulse indeed begins at $P_m \approx (\alpha - \kappa_0 - \gamma)/\kappa_0$ and continues until P_m becomes much larger than unity. At the end of the generation, when P_m again becomes of the order of unity, a certain compression of the pulse again occurs. This agrees well with the dependence of the pulse compression rate per pass on the pulse intensity P_m at the maximum, obtained in[8]:

$$W = \frac{d\tau}{dk} = -\tau \frac{\varkappa_0 P_k}{(1+P_k)(2+P_k)}. \tag{23}$$

It follows from (23) that the rate of compression of the pulse is maximal at $P_k = \sqrt{2}$ and tends to zero in limiting cases of small and large intensities.

Second, it is seen from Fig. 1b that when the pulse duration becomes of the order of T_{1b}, the pulse becomes asymmetrical and the leading front becomes much steeper than the trailing edge. At the end of the generation the pulse duration is $\tau \approx 0.5\,T_{1b}$. In principle, even shorter pulse durations can be generated. To this end it is necessary that the attainable minimum pulse duration τ_{min}, which is reached in a laser with a non-inertial absorber, be much shorter than the relaxation time T_{1b} of the real absorber used in the laser. Figure 2a shows an example of the development of generation in such a case. In this example, the pulse duration reaches a value $\sim 0.2\,T_{1b}$ already in the center of the generation and subsequently remains unchanged. The waveform of the pulse $P(t)$ and the corresponding change of the absorption $\kappa(t)$ are shown in Fig. 2b.

The influence of the absorber on the evolution of the pulse in the region where the duration of the pulse is small ($\tau \ll T_{1b}$) and the power is large ($P_m \gg 1$) can be understood by considering the change of the absorption $\kappa(t)$ under the influence of such a pulse. The change of the absorption during the time of passage of the pulse can obviously be considered by neglecting the relaxation. Then the decrease of the absorption is described by the expression

$$\varkappa_1(t) = \varkappa_0 \exp\left\{-\frac{1}{T}\int_{-\infty}^{t} P(t')\,dt'\right\}. \tag{24}$$

Tha absorption decreases to a value $\kappa_{min} \approx \kappa_0 \exp \times (-\tau P_m/T_{1b})$ within a time $\tau' \approx T_{1b}/P_m$. After passage of the pulse, the absorption relaxes to the initial state within a time of the order of T_{1b}, practically independently of the pulse:

$$\varkappa_2(t) = \varkappa_{min} \exp(-t/T_{1b}) + \varkappa_0[1 - \exp(-t/T_{1b})]. \tag{25}$$

The "transmission" time of the absorber is proportional to the power of the pulse and can therefore be quite small. The "blocking" time of the absorber does not depend on the power and is determined by the relaxation time T_{1b}. Therefore the slope of the leading edge of the pulse can be much larger than that of the trailing edge. The leading edge, in principle, can become steplike, but in practice this does not happen in a laser, since saturation of the gain causes the pulse power to decrease quite rapidly. However, the duration of the leading edge can be shorter by one order of magnitude than the relaxation time T_{1b}.[3)]

It is now easy to understand how the dispersion of the medium inside the resonator affects the evolution of the pulses in a laser operating in a nonstationary regime. During those stages of pulse development when its duration is large or when there is a noticeable rate of duration compression, the dispersion obviously does not influence the pulse development. However, in the region $P_m \gg 1$, when the pulse duration is small and the pulse compression rate is zero, broadening of the pulse is possible. This broadening is described approximately by the expression

$$\tau \approx \tau_m \left\{1+\left[\frac{2kL}{\tau_m{}^2 c}\left(\frac{\partial n}{\partial \omega}+\frac{\omega_0}{2}\frac{\partial^2 n}{\partial \omega^2}\right)\right]^2\right\}^{1/2}, \qquad (26)$$

where τ_m is the pulse duration at the instant when the compression stops, k is the number of passes reckoned from the instant of the stopping of the compression, $Ln(\omega)$ is the total optical path of the dispersive medium in the resonator. The main dispersive element is usually the glass or the crystal of the active medium. At length L = 10 cm and a dispersoin $\partial n/\partial\omega \approx 10^{-17}$ sec^{-1}, for example, the broadening after 30 passes will be influential only for pulses with duration $\tau_m \approx 5 \times 10^{-13}$ sec.

7. REMARK CONCERNING THE RELAXATION OF A SATURABLE SOLUTION

A "fast" saturable solution of a dye used in ultrashort-pulse laser is usually regarded as a two-level system in which the molecules at the upper level have a short effective lifetime T_{1b}, determined by radiative and nonradiative transitions to the ground state. The probability of the radiative relaxation of the excited level for allowed transitions with an oscillator strength $f \approx 1$ in the visible band does not exceed 10^9 sec^{-1}. Nonradiative relaxation may greatly reduce the lifetime of the excited molecules. However, the rate of nonradiative degradation of the excitation cannot reach the limiting values 10^{12}–10^{13} sec^{-1}, since kT (T—temperature of the solution) is much smaller than the excitation energy of the molecule.

Nonetheless, relaxation rates of the saturated state of the solution on the order of 10^{13} sec^{-1} can be reached in practice. Besides the excited singlet state S_2, into which the molecule goes over from the ground state S_0, the molecule can have a low-lying electronic state S_1 (Fig. 3). All these states have also a set of vibrational levels. The excited molecules in the state S_2 relax as the result of the nonradiative transitions to lower levels of the excited state S_1. The rate of such a relaxation, called internal conversion, is quite large and, according to[24] (see also the reviews[25,26]) amounts to $T_{12}^{-1} = 10^{10}$–10^{13} sec^{-1}. The molecules return from the lower excited state S_1 to the ground state much more slowly, within a time $T_1 \gg T_{12}$. As the result of the fast transitions of the molecules between the excited levels, the saturated state of the solution relaxes to an absorbing state (but not the initial state!) with absorption $\kappa_0/2$ within a characteristic time T_{12}, and then, as the result of the transition of the molecules to the ground state, the solution relaxes to the initial state with absorption κ_0 within the much longer time T_1. A saturable solution with such a two-step relaxation is quite suitable for the production of ultrashort pulses[4)]. The first relaxation ensures compression of the pulses to a value $\sim T_{12}$, and the second relaxation returns the solution to the initial state prior to the next arrival of the pulse ($T_1 \ll T$). Pulse compression in a laser with such a saturable solution is similar in many respects to the picture considered above. A more detailed consideration of this question will be presented later.

CONCLUSION

We have developed in the article the theory of generation of ultrashort pulses in a laser with a nonlinear absorber, including all phases of generation development. It follows from the theory that the limiting duration of ultrashort pulses, determined by the width of the amplification band, can be realized in practice. To this end, it is proposed to eliminate the broadening of the pulse in the linear generation phase and to use saturable solutions with two-step relaxation of the saturated state of the molecules. The last phase of development of the generation in a laser for extremely short light pulses, consisting in formation of a pulse of limiting stationary duration, is not described by the present theory. It calls for an account of the shape of the amplification line. This question will be dealt with in a later article. Apparently it is necessary to take into account also the transverse structure of the pulses, for at a duration of 10^{-12} sec the transverse dimension of the pulse becomes appreciably larger than the longitudinal one, i.e., the pulse has in space the form of a strongly flattened electromagnetic bunch.

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3) We note that if the transverse relaxation of the molecules in the solution is shorter than T_{1b} by not more than one order of magnitude, than the initial rate equation (3b) becomes incorrect in this case. It must be replaced by the equations for the polarization density $\mathscr{P}_b(t)$ and the population $N_b(t)$ of the absorbing medium. As a result, the response of the absorber to a powerful pulse with duration $\sim T_{2b}$ becomes oscillatory. The resulting absorption oscillations of the $n\pi$-pulse type, generally speaking, can lead to oscillations in the pulse [23]. However, this does not occur here, owing to the presence of gain $\alpha \gtrsim \kappa$ in the laser, which increases the intensity of the pulse and by the same token prevents accumulation of the distortions of its wave form. The oscillations in the pulse can appear only when $\alpha \ll \kappa$, a case not realized in the laser considered here.

[1] A. J. De-Maria, D. A. Stetser, and H. Heynau, Appl. Phys. Lett. 8, 174 (1966).

[2] A. J. De-Maria, D. A. Stetser, and W. H. Glenn, Jr. Science, 156, 1557 (1967).

4)It is not excluded that this mechanism takes place in polymethine, with the aid of which it is possible to obtain ultrashort pulses of duration 10^{-12} sec [27,28].

[3] J. A. Armstrong, Appl. Phys. Lett. 10, 16 (1967); W. H. Glenn and M. J. Brienza, Appl. Phys. Lett. 10, 221 (1967).
[4] J. A. Giodmaine, P. M. Rentzepis, S. L. Shapiro, and K. W. Wecht, Appl. Phys. Lett. 11, 216 (1967); P. M. Rentzepis and M. A. Duguay, Appl. Phys. Lett. 11, 218 (1967).
[5] T. I. Kuznetsova, V. I. Malyshev, and A. S. Markin, Zh. Eksp. Teor. Fiz. 52, 438 (1967) [Sov. Phys.-JETP 25, 286 (1967)].
[6] E. Garmire and A. Yariv, IEEE J. Quantum Electronics, QE-3, 222 (1967).
[7] T. I. Kuznetsova, FIAN Preprint No. 25, 1967; Abstracts of Third All-union Conference on Nonlinear Optics, Erevan, 1967.
[8] V. S. Letokhov, ZhETF Pis. Red. 7, 35 (1968) [JETP Lett. 7, 25 (1968)].
[9] P. P. Sorokin, J. J. Luzzi, J. R. Lankard, and G. D. Pettit, IBM J. Res. and Develop. 8, 182 (1964).
[10] G. Bret and F. Gires, Appl. Phys. Lett. 4, 175 (1964).
[11] V. L. Borovich, V. S. Zuev, and V. A. Shcheglov, Zh. Eksp. Teor. Fiz. 49, 1031 (1965) [Sov. Phys.-JETP 22, 717 (1966)].
[12] V. I. Malyshev, A. S. Markin, and A. A. Sychev, ZhETF Pis. Red. 6, 503 (1967) [JETP Lett. 6, 34 (1967)].
[13] V. S. Letokhov and V. N. Morozov, Zh. Eksp. Teor. Fiz. 52, 1296 (1967) [Sov. Phys.-JETP 25, 862 (1967)].
[14] V. S. Letokhov, ibid. 54, 1392 (1968) [27, 746 (1968)].
[15] V. I. Bespalov, Izv. vuzov, Radiofizika 11, 320 (1968).
[16] L. Mandel and E. Wolf. Rev. Mod. Phys. 37, 231 (1965).
[17] V. S. Letokhov, Zh. Eksp. Teor. Fiz. 50, 765 (1966) [Sov. Phys.-JETP 23, 506 (1966)]; FIAN Preprint A-45, 1965.
[18] M. A. Duguay, S. L. Shapiro, and P. M. Rentzepis, Phys. Rev. Lett. 19, 1014 (1967); S. L. Shapiro, M. A. Duguay, and L. B. Kreuzer, Appl. Phys. Lett. 12, 36 (1968).
[19] W. R. Sooy, Appl. Phys. Lett. 7, 36 (1965).
[20] V. I. Malyshev, A. S. Markin, A. V. Masalov, and A. A. Sychev, FIAN Preprint No. 34, 1968; Zh. prikl. spektroskop. (in press).
[21] V. S. Zuev, and V. A. Shcheglov, Zh. prikl. spectroskop. 5, 604 (1966).
[22] V. E. Khartsiev, D. I. Stasel'ko, and V. M. Ovchinnikov, Zh. Eksp. Teor. Fiz. 52, 1457 (1967) [Sov. Phys.-JETP 25, 965 (1967)].
[23] S. L. McCall and E. L. Hahn, Phys. Rev. Lett. 18, 908 (1967).
[24] M. Kasha, Disc. Farad. Soc. 9, 14 (1959).
[25] S. K. Lower and M. A. El-Sayed, Chem. Rev. 66, 199 (1966).
[26] V. L. Ermolaev, Usp. Fiz. Nauk 80, 3 (1963) [Sov. Phys. Usp. 6, 333 (1963)].
[27] N. G. Basov, P. G. Kryukov, V. S. Letokhov, and Yu. V. Senatskiĭ, Paper at Fifth Internat. Conf. on Quantum Electronics, Miami, 1968.
[28] S. D. Kaĭtmazov, I. K. Krasyuk, P. P. Pashinin, and A. M. Prokhorov, Dokl. Akad. Nauk SSSR 180, 1331 (1968) [Sov. Phys.-Dokl. 13, 591 (1968)].

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