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# GENERATION OF ULTRASHORT LIGHT PULSES BY MEANS OF LASERS

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A review is given of the theory of the generation of ultrashort light pulses in lasers with bleachable filters. Attention is mainly concentrated on the statistical aspects of the formation of ultrashort light pulses in such lasers. The topics discussed include the formation of laser modes, the narrowing of the spectrum in the stage before the bleaching of the filter, the transformation of the field profile during the bleaching process, the statistics of the appearance of single ultrashort pulses, and the influence of the gain saturation on the time characteristics of the radiation. Several additional effects, which influence the final structure of the radiation, are considered. A quantitative theory is given of the widely used two-photon method for measuring ultrashort pulses and a basic shortcoming of this method is pointed out: it is not possible to find whether one or many pulses appear in a given period.

## CONTENTS

1.	Introduction	25
2.	Successive Stages of the Development of Stimulated Emission in a Laser With a Bleachable	
	Filter	26
3.	Subthreshold Establishment of a Quasiperiodic Fluctuation Pattern (Mode Formation	
	Process	27
4.	Narrowing of the Emission Spectrum in the Stage Before the Bleaching of a Filter	29
5.	Transformation of the Field Profile in the Filter Bleaching Process	31
6.	Statistics of the Appearance of Ultrashort Pulses During Filter Bleaching	33
7.	Influence of Gain Saturation on the Final Field Profile	36
8.	Additional Effects Influencing the Final Structure of the Radiation	37
9.	Two-Photon Method for Recording the Time Structure of the Radiation	39
10.	Conclusions	42
Lite	erature Cited	43

# 1. INTRODUCTION

IN 1966 several workers<sup>[1-3]</sup> reported the generation of laser radiation with remarkable properties. A time scan of the intensity of this radiation was a periodic sequence of pulses whose duration was considerably less than the separation between them. By way of illustration, Fig. 1 shows an oscillogram of the intensity of the radiation emitted by a neodymium laser, as reported in<sup>[4]</sup>. Here, the pulse repetition period is equal to the time required for a round trip along the resonator and the duration of the pulses—amounting to  $3 \times 10^{-9}$  sec is determined by the width of the spectrum. The instantaneous values of the pulsed power are several times larger than the average radiation power.

Radiation of this type is obtained when a resonator contains a bleachable filter, which is a substance whose transmission increases with increasing radiation intensity. Clearly, the concentration of the radiation in short bursts reduces the absorption of light in such a substance and, therefore, such concentration is energetically more favorable than the emission of radiation which is continuous in time.

These qualitative considerations and the first successful experiments seemed extremely promising. It was hoped to achieve generation of extremely short pulses and thus attain very high instantaneous powers by the simple increase of the width of the generated spectrum.



FIG. 1. Oscillogram of a giant pulse emitted by a neodymium laser. [4] The pulse repetition period is 22 nsec and the pulse duration ( $\sim$ 3 nsec) is equal to the reciprocal of the width of the spectrum.

In fact, considerable difficulties were encountered whose significance was not understood immediately. The position was complicated by the fact that erroneous methods for measuring the time characteristics of the laser radiation were put forward in 1967 and found wide application. These methods were used by many workers to conclude that they produced very short pulses and reached record power levels.

Only the recent and more careful investigations of the generation of ultrashort pulses established that regular sequences of  $\sim 10^{-11}$  sec pulses were produced. This

25

was established reliably by the use of unique apparatus in which the time characteristics of the radiation were recorded directly (see<sup>[5]</sup>).

The present review presents a quantitative theory which describes the whole process of the generation of radiation in a laser with a bleachable filter. We shall show how a regular sequence of pulses can be produced in a laser without any external modulation.<sup>1)</sup> We shall discuss specially the statistical nature of the action of such a laser: the appearance of a regular time pattern in the output radiation is characterized by some probability. We shall show how the parameters of a laser should be varied in order to increase the probability of the appearance of a periodic sequence of single ultrashort pulses. In Chap. 2 we shall give a qualitative description of the successive stages of the development of stimulated emission in a laser with a bleachable filter. The results of a quantitative analysis of the successive stages of the operation of a laser of this type are given in Chaps. 3-7.

Ultrashort light pulses have a wide range of applications in technology and physical investigations. For example, they can be used in studies of the properties of matter and vacuum in extremely strong electromagnetic fields, in measurements of the lifetimes of molecular systems ( $\sim 10^{-12}$  sec), and in initiation of thermonuclear reactions as a result of heating of matter by laser radiation. The possible applications of ultrashort light pulses have been discussed widely in the literature (see, for example, reviews given in<sup>[6,7,51,52</sup>]) and we shall not consider them here.

#### 2. SUCCESSIVE STAGES OF THE DEVELOPMENT OF STIMULATED EMISSION IN A LASER WITH A BLEACHABLE FILTER

In the present chapter we shall give a quantitative description of the successive stages of the development of stimulated emission in a laser with a bleachable filter. This description will also serve as a summary of the content of the chapters in the present review which give a quantitative discussion of the processes occurring at different stages in the appearance of coherent radiation.

The operation of a solid laser under giant pulse conditions begins at the moment of switching-on the pulsed pumping whose action transfers active atoms to a higher energy level. At the beginning of a pumping pulse the intensity of the radiation at the frequency of the laser transition is zero and the active medium shows practically no gain at this frequency. Next, the atoms excited by the pumping radiation begin to emit spontaneous photons at the laser transition frequency. The spectrum of the spontaneous radiation is identical with the spectrum of the luminescence line corresponding to the laser transition. Some of the spontaneously emitted photons are reflected by the resonator mirrors and they return to the active substance where they are amplified. However, as long as the population of the atoms at the upper excited level is considerably less than the threshold value, the fraction of these reflected and amplified photons is less than the fraction of the photons generated directly in the spontaneous emission process. Since the spontaneous emission is essentially a fluctuation process, the intensity of the radiation fluctuates at a characteristic correlation time  $\tau_{\rm corr} \sim 1/\Delta \omega_{\rm lum}$ .

When the population of the upper active level approaches the threshold value, the fraction of the reflected photons increases and this amplifies partly the noise radiation which is emitted by the active medium and reflected by the resonator mirrors. At the moment when the contribution of the amplified noise exceeds the contribution of the direct spontaneous radiation we find that the fields become periodically correlated at the moments t and t + T, where T is the time taken by radiation to travel across the resonator in both directions. This round-trip time is determined by the geometry of the resonator and, in the simplest case of a linear resonator with a standing wave, it is given by T = 2L/c, where L is the distance between the mirrors.

The total width of the spectrum is still ~  $\Delta\omega_{lum}$  (the correlation time is short:  $\tau_{corr} \sim 1/\Delta\omega_{lum}$ ). However, the presence of a periodic correlation corresponds to in the spectroscopic language—the formation (under the influence of the resonator) of single discrete modes forming an equidistant set of frequencies  $\omega_{\bf k} = \omega_0$ + k( $2\pi/T$ ). We must stress that a quasiperiodic fluctuation pattern is formed, generally speaking, before the laser threshold is reached. These initial fluctuations are important in the formation of the final time pattern of the radiation.

The gain of the active medium at a frequency  $\omega_k$  is proportional to the product of the difference of the populations of the active levels and the function  $g(\omega_k)$ , which describes the spectral profile of the luminescence line. Therefore, during the growth of the inversion under the influence of the pumping radiation, the threshold conditions (the excess of the gain over the losses) are first satisfied by the central frequency and then by other frequencies. The process of regenerative amplification of the noise transforms in a continuous manner into the emission of coherent radiation. Under these conditions, the frequencies close to the center of the gain profile are amplified preferentially.

At this stage, the laser is a linearly amplifying system because the field intensity is still low and the field does not give rise to nonlinear effects in the filter or in the active substance. This linear amplification stage lasts until the onset of bleaching of the filter. Since the intensity of the radiation at the onset of the bleaching (increase in transparency) stage is many orders of magnitude higher than the intensity of the spontaneous radiation emitted by the active medium, this linear amplification stage is necessarily quite long. Therefore, the narrowing of the emission spectrum as a result of the inhomogeneous distribution of the gain over the spectrum is very strong.

Thus, at the moment when the nonlinear stage begins (the stage of bleaching of the filter) the radiation has a quasiperiodic noise pattern and the width of the spectrum is  $\Delta \omega_1$ , which is much less than the initial width of the luminescence line  $\Delta \omega_{lum}$ . During one period there

<sup>&</sup>lt;sup>1)</sup> Periodic sequences of pulses had been generated in lasers whose Q factor was switched externally (see, for example, [<sup>50</sup>]) before the appearance of papers describing lasers with bleachable filters. [<sup>1-3</sup>] There were many great technical difficulties which impeded widespread use of lasers with external Q-switching. However, the operation of such lasers is clearly understood.

are  $\sim \Delta \omega_1 T$  random amplitude peaks. Since the initial spontaneous noise is amplified linearly, the field at the beginning of the bleaching stage is a complex Gaussian random quantity.

The field profile is modified during the bleaching stage. This modification consists in the preferential amplification of those parts of the profile at which the intensity has its highest value. Multiple passage of the radiation through the bleachable filter reduces the duration of each intensity peak. At the same time, the strongest peaks are amplified more rapidly than the weaker peaks. This gives rise to a time distribution of the intensity which is much more strongly inhomogeneous than that in the initial Gaussian process. We may find that all the laser radiation energy in a period is concentrated in one or several very strong peaks. The formation of such a time distribution is the most important feature of the operation of a laser with a bleachable filter. The time distribution with one strong peak in a period can be achieved only with a certain probability because the field before bleaching is a random function of time. This probability is calculated in Chap. 6 as a function of the laser parameters.

The next stage is a rapid amplification (the filter is now fully bleached and completely transparent) of the field profile formed during the bleaching stage. The nonlinearity of the gain in the active substance becomes gradually more pronounced. The gain saturation determines the amplitude of the giant pulse and the time during which the light energy stored in the resonator is emitted.

This schematic description of the operation of a laser with a bleachable filter is based on the assumption that the various stages are divided by sharp boundaries. Actually, there is no absolutely sharp boundary between the end of one stage and the beginning of the next. For example, the frequency dependence of the gain of the active medium is important at all stages. However, this frequency is most important in the linear stage before bleaching (this stage is longest) and its influence during other stages can be ignored.

The aforementioned effects which influence the operation of a laser with a bleachable filter are basic in the sense that they must appear in all lasers of this type. This is used to construct a quantitative theory of the phenomena which occur at different stages of the development of stimulated emission: this theory is given in Chaps. 3-7.

Apart from the effects described in the preceding paragraphs, several others may exert a considerable influence on the final field profile. Their influence depends on the actual parameters of a laser. Some of these effects are listed below.

The frequency dependence of the refractive index of the active medium (the dispersion of glass or ruby) results in a non-equidistant mode pattern which is known as the dispersion blurring of the pulses in time. The pulses retain their spectral width and acquire phase modulation.

An increase in the refractive index of glass or ruby under the influence of the strong field of the laser radiation does not, in the first approximation, affect the pulse duration but it does broaden the spectrum and gives rise to phase modulation. When the radiation intensity is high, various nonlinear losses impair the conditions for the amplification of the strongest peaks. Consequently, the energy may be spread over a larger number of weaker peaks.

These "additional" effects are considered in Chap. 8 (they may be extremely important in specific situations).

In view of the wide use of the two-photon method for measuring ultrashort pulses it has seemed desirable to consider this method in some detail. Chapter 9 gives a brief description of this method and points out its basic shortcoming: it is not possible to determine the instantaneous power by this method.

## 3. SUBTHRESHOLD ESTABLISHMENT OF A QUASIPERIODIC FLUCTUATION PATTERN (MODE FORMATION PROCESS)

At the beginning of the whole process the gain of the active medium is practically zero and the field perturbations in the resonator decay in accordance with the law  $\mathscr{E} \propto \{-\alpha t/2\}$ , where  $\alpha$  is the decay constant. The constant  $\alpha$  includes various losses in the laser: the losses due to the reflection from the mirrors, as well as those due to the linear scattering and absorption in the filter (at this stage the filter is a linear element). The attenuation of the field after a one-way trip through the resonator is given by  $\exp\{-\alpha T/2\}$ . Under typical conditions the product  $\alpha T$  ranges from 0.1 to 3-5. (For example, in the case of 50% losses at each of the mirrors and  $\sim 20\%$  transmission of the filter in the unbleached state, the product  $\alpha T$  is 3.0.) We shall consider a laser emitting radiation modes with the lowest transverse index. The spectrum of such modes can be regarded as equidistant and the mode spacing is

$$\Omega = 2\pi/T. \tag{3.1}$$

The problem of establishment of the mode pattern must be considered bearing the following point in mind. Because of the decay of the radiation, the spectral width  $\Gamma$  of the field of some mode is  $\Gamma \approx \alpha$ . In many cases, this width is of the same order as the mode spacing  $\Omega = 2\pi/T$ . In this situation, the concept of a mode is not equivalent to the concept of a spectral component: the field of the k-th mode has a continuous spectrum of frequencies which are overlapped significantly by the spectra of the fields of the k ± 1-th, k ± 2-th, and other modes. In view of this situation, we shall consider in detail the establishment of modes under the action of spontaneous radiation during gradual amplification.

We shall consider the simplest model of a laser in which the gain and the losses are distributed uniformly over the length of the resonator.<sup>2)</sup> The field in the resonator can be represented as an expansion consisting of a complete set of functions

$$E(z, t) = e^{-i\omega_0 t} \sum_{k=-\infty}^{+\infty} \cos\left[\left(p_0 + k\frac{\Omega}{c}\right)z\right] E_k(t).$$
(3.2)

We can see that  $\omega$ , which is the central frequency of the luminescence line of the active medium, is clearly distinguished and that  $p_0 = \omega_0/c$ . The sources of spontaneous noise (atoms) are much smaller than the wavelength

<sup>&</sup>lt;sup>2)</sup>This simple laser model is used only in the present chapter; it will not be needed in subsequent chapters.

and are distributed uniformly over the length of the resonator. The corresponding noise polarization  $P_n(z, t)$  is  $\delta$ -correlated with respect to z:

$$\langle P_n^*(z, t) P_n(z', t') \rangle = g(t, t') \,\delta(z - z').$$
 (3.3)

A truncated system of equations for the amplitudes of the spatial modes  $\mathbf{E}_{\mathbf{k}}(t)$  is of the form

$$\dot{E}_{k}(t) + \frac{1}{2} (\alpha - \hat{\beta}(t)) E_{k}(t) - ik\Omega E_{k}(t) = f_{k}(t).$$
 (3.4)

We shall now explain the notation used in the above equation. The factor  $\hat{\beta}(t)$  describes the amplification of the field of the k-th spatial mode of the active medium and the gain increases with time t under the action of the pumping pulse. Strictly speaking, the gain band  $(\Delta \omega = \Delta \omega_{lum})$  is of finite width and, therefore, the term describing the amplification of the field  $E_k(t)$  should be of the form  $\int_{-\infty}^{t} \beta(t, t') E_k(t') dt'$ . The interval |t - t'| in which the kernel of the integral eccenter  $\theta(t, t')$  differe

which the kernel of the integral operator  $\beta(t, t')$  differs significantly from zero is of the order of  $\Delta t$ 

~  $(\Delta \omega_{lum})^{-1}$ . We shall assume (and confirm the validity of this assumption by the results of subsequent calculations) that the field of the k-th spatial mode depends on time in accordance with the law  $E_{k}(t) = e^{-ik\Omega t}E_{k,slow}(t)$ , where the function  $E_{k,slow}(t)$  varies only in time intervals considerably greater than  $(\Delta \omega_{lum})^{-1}$ . In this case, the effect of the operator  $\beta(t)$  reduces practically to multiplication by the number  $\beta_{\mathbf{k}}$ , which is a function of the frequency  $k\Omega$ . This number is real (it has only the active component) at the center of the luminescence line (k = 0). By virtue of the Kramers-Kronig relationships, the imaginary component of the number  $\beta_k$  near the center of the line is proportional to k: Im  $\beta_{\mathbf{k}} = \Lambda \mathbf{k} \Omega$ . The imaginary component Im  $\beta_{\mathbf{k}} \sim \mathbf{k}$  can be eliminated by redefining the mode spacing as  $\Omega' = \Omega + \frac{1}{2}\Lambda \Omega$ . This corresponds to an allowance for the change in the group velocity of the radiation in the active medium as a result of a resonance transition. We shall assume that the values of  $\beta_k$  are real for those modes which are located near the center of the line. When this redefinition is adopted, we find that the numbers  $\beta_k$  contain imaginary components proportional to  $(k \Omega / \Delta \omega_{lum})^3$ ; we shall drop these imaginary corrections from the final formulas because they are small for the strongest central modes.

The right-hand part of Eq. (3.4) contains the k-th spatial component of the noise polarization  $f_k(t) \sim \int P_n(z, t) \cos[(p_0 + k\Omega/c)z] dz$ , which is a random function of time.<sup>3)</sup> Since the distribution of the noise sources over the length of the resonator is uniform and the system of functions  $\cos[(p_0 + k\Omega/c)z]$  is orthogonal, the noise sources corresponding to different spatial modes are independent:

$$\langle f_{k_2}^*(t') f_{k_1}(t) \rangle = \delta_{k_1 k_2} H(t) \int_{-\infty}^{+\infty} G(\omega) e^{-i\omega(t-t)'} d\omega.$$
 (3.5)

Consequently, the amplitudes  $\mathbf{E}_{\mathbf{k}}$  of the various spatial modes are also independent. The last equation is derived using the Wiener-Khinchin theorem on the relationship between the correlation function and the power

spectrum;  $G(\omega)$  is the normalized profile of the luminescence line; the slowly varying function H(t) represents the gradual rise in the number of atoms at the upper excited level of the active transition under the action of the pumping pulse.

The solution of Eq. (3.4) is of the form

$$E_{k}(t) = \int_{-\infty}^{t} dt' f_{k}(t') \exp\left\{\int_{t'}^{t} \left[-ik\Omega + \frac{1}{2}\beta_{k}(t'') - \frac{1}{2}\alpha\right] dt''\right\}.$$
 (3.6)

The average of the fluctuation ensemble is  $\langle E_{\mathbf{k}}(t) \rangle = 0$ . We shall now determine the correlation function  $\langle E_{\mathbf{k}}^{*}(t)E_{\mathbf{k}}(t + \tau) \rangle$ . This function is given by

$$= \int_{-\infty}^{t} dt_{1} \int_{-\infty}^{t+\tau} dt_{2} \langle f_{k}^{*}(t_{1}) f_{k}(t_{2}) \rangle \exp \left\{ \int_{t_{1}}^{t} \left[ ik\Omega + \frac{1}{2} \beta_{k}^{*}(t') - \frac{1}{2} \alpha \right] dt' + \int_{t_{2}}^{t+\tau} \left[ -ik\Omega + \frac{1}{2} \beta_{k}(t'') - \frac{1}{2} \alpha \right] dt'' \right\} . (3.7)$$

Substituting Eq. (3.5) into Eq. (3.7) and bearing in mind that the width of the luminescence line  $\Delta \omega_{\text{lum}}$  of all the solid lasers is much greater than any of the quantities  $\alpha$ ,  $\beta$ , or  $\Omega$ , we obtain

$$\langle E_{k}^{*}(t) E_{k}(t+\tau) \rangle = e^{-ik\Omega\tau} e^{-\frac{1}{2} (\alpha - \beta_{k}(t)) |\tau|} \frac{2\pi H(t) G(k\Omega)}{\alpha - \beta_{k}(t)}.$$
 (3.8)

The above equation is derived on the assumption that the gain  $\beta_{\mathbf{k}}(t)$  varies sufficiently slowly in time. Equation (3.8) describes the following two effects.

First of all, it follows from this equation that the intensity of the noise oscillations is highest at the center of the luminescence line (the factor  $G(k \Omega)[\alpha - \beta_k(t)]^{-1}$ ). According to this formula, the number of the most effectively excited spatial modes should decrease without limit when the laser threshold,  $\beta_0(t_{th}) = \alpha$ , is approached. A more accurate analysis of this effect is given in Chap. 4.

Moreover, it follows from Eq. (3.8) that the width of the <u>frequency</u> spectrum of the amplitude of the k-th <u>spatial</u> mode becomes  $\Gamma_{\mathbf{k}} \approx \alpha - \beta_{\mathbf{k}}(t)$ . When the gain approaches the threshold value, an increasing number of modes satisfies this condition

$$\Gamma_k \approx \alpha - \beta_k(t) \ll \Omega. \tag{3.9}$$

This means that the spectrum of fluctuations of the amplitude of the k-th mode has practically no overlap with the spectra of the fluctuations of the (k - 1)-th and (k + 1)-th modes. Therefore, when the threshold is approached sufficiently closely, the concepts of the spatial mode and of the spectral component in the equidistant set of frequencies become practically identical. This is the main conclusion which follows from the analysis given in the present chapter.

In the next chapter (Chap. 4) we shall study the evolution of the distribution of the energy over the various spatial modes. In view of what we have just said, this will be equivalent to the calculation of the spectrum of the electromagnetic field in the laser.

The radiation field emerging from the laser (for example, the field at the point z = 0) is given by the sum of the contributions of the various modes:

$$E(z=0, t) = \sum E_h(t).$$
 (3.10)

If we use the fact that the amplitudes of different modes are independent, we can find quite easily the expression

<sup>&</sup>lt;sup>3)</sup>A consistent allowance for the quantum-mechanical noise of spontaneous radiation is discussed in [<sup>8</sup>]. We shall not stress explicitly the quantum nature of the quantities considered but all the calculations given here apply also to the quantum treatment.

for the correlation function  $\langle \mathbf{E}^*(t)\mathbf{E}(t + \tau)\rangle$ . We shall give an explicit expression for the case in which the preferential growth of the energy of the central modes becomes significant, i.e., when the spectrum of the effectively excited modes becomes two or three times narrower than the luminescence line  $[\Delta \omega_{\text{exc}} \leq (1/3) \Delta \omega_{\text{lum}}]$ :

(0, 1)

$$\langle E^{\bullet}(t) E(t+\tau) \rangle = \exp\left\{-\frac{1}{2}(\alpha - \beta_0(t)) T \left| \left[\frac{\tau}{T}\right] \right| \right\} R(\tau), \quad (3.11)$$

$$R(\tau) = \sum_{k=-\infty}^{+\infty} \frac{2\pi H(t) G(k\Omega)}{\alpha - \beta_k(t)} e^{-ik\Omega\tau}.$$
 (3.12)

The expression  $[\tau/\mathbf{T}]$  in Eq. (3.11) represents the integral part of the ratio  $\tau/\mathbf{T}$  and the function  $\mathbf{R}(\tau)$  has a period T. The function  $\mathbf{R}(\tau)$  describes the correlation of the fields in one period and differs considerably from zero in the interval  $|\tau| \leq (\Delta \omega_{\text{exc}})^{-1}$ . Equation (3.11) describes the appearance of correlation between the fields at moments separated by integral numbers of periods: this means that a quasiperiodic fluctuation pattern is established and the decay of the correlation in one period is represented by the factor  $\exp\{-\frac{1}{2}[\alpha - \beta_0(t)]T\}$ .

### 4. NARROWING OF THE EMISSION SPECTRUM IN THE STAGE BEFORE THE BLEACHING OF A FILTER

In this chapter we shall consider the linear stage of the development of stimulated emission up to the moment of bleaching of the filter. The characteristic features of this stage were first discussed by  $\text{Sooy}^{[11]}$  and in greater detail by Sushchik and Freidman.<sup>[12]</sup> We shall concentrate mainly on the calculation of the following quantities, which characterize the linear stage: 1) the time of rise of the intensity of the optical oscillations to the value at which the nonlinearity of the bleachable filter becomes important; 2) the number of modes which are effectively excited at the onset of the bleaching process; 3) the gain  $\beta(t_1)$  which is established at that moment. The last two quantities will be used to describe the operation of the laser in the subsequent stages.

We have pointed out in Chap. 3 that, beginning from a certain moment, the development of coherent radiation from continuous noise can be described by the fields of various modes. The equation for the averaged (over the fluctuation ensemble) energy of the k-th mode  $u_k(t)$ -expressed in ergs—is of the form

$$\frac{du_{k}(t)}{dt} - (\beta_{k}(t) - \alpha_{k}) u_{k}(t) = \hbar \omega_{0} \frac{N_{2}}{N_{2} - (N_{1}g_{2}/g_{1})} \beta_{k}(t); \qquad (4.1)$$

here,  $\hbar\omega_0$  is the average value of the energy of a photon corresponding to the active transition frequency;  $N_2$  and  $N_1$  are the populations of the upper and lower laser levels;  $g_2$  and  $g_1$  are the statistical weights of these levels.

In the linear stage the number of effectively excited modes decreases considerably. Therefore, we can expand the coefficients  $\alpha_k$  and  $\beta_k(t)$  in powers of  $k \Omega/\Delta \omega$ . Near the center of the luminescence line these quantities can be represented in the form

$$\beta_{k}(t) = \beta(t) \left( 1 - \frac{(k\Omega)^{2}}{(\Delta \omega_{\text{lum}})^{2}} \right), \qquad (4.2)$$

$$\alpha_{k} = \alpha_{nl} + \alpha_{1} \left( 1 + \frac{(K\Omega)^{2}}{(\Delta \omega_{res})^{2}} \right)$$
 (4.3)

We shall assume that the linear losses in the resonator  $(\alpha_l)$  can also depend on the frequency and are minimal at the center of the line. On the other hand, the non-linear losses in the filter  $(\alpha_n l)$  will be assumed to be independent of the frequency.

At some moment  $t_{th}$  the losses and the gain become equal at the central frequency  $k\Omega = 0$ :

$$\beta(t_{\rm th}) = \alpha_{\rm nl} + \alpha_{\rm l} \equiv \alpha. \tag{4.4}$$

For typical laser systems the value of  $T_{pump}$ , which is the time in which the gain changes significantly under the influence of pumping, amounts to  $T_{pump} \sim 10^{-3}-10^{-4}$  sec, whereas the duration of the linear stage of the development of stimulated emission is  $10^{-5}-10^{-6}$  sec. Therefore, near  $t = t_{th}$ , where the phenomena of interest to us are taking place, we may assume that the gain is a linear function of time:

$$\beta(t) = \beta(t_{\rm th}) + \beta(t_{\rm th})(t - t_{\rm th})$$
(4.5)

[from now onward we shall drop the argument of  $\dot{\beta}(t_{th})$ ], so that after substitution of Eqs. (4.3)-(4.5) the coefficient  $\beta_k(t) - \alpha_k$  in Eq. (4.1) becomes

$$\beta_k(t) - \alpha_k = (t - t_{\text{th}}) \dot{\beta} k^2 \Omega^2 \left[ \frac{\alpha + \dot{\beta} (t - t_{\text{th}})}{(\Delta \omega_{\text{lum}})^2} + \frac{\alpha_1}{(\Delta \omega_{\text{peg}})^2} \right].$$
(4.6)

The change in  $\beta(t)$  throughout the linear stage is quite small (for the same reasons):  $(t_1 - t_{th})\hat{\beta} \ll \beta(t_{th}) = \alpha$ . Therefore, we can ignore the product  $k^2 \Omega^2 (t - t_{th})\hat{\beta}$  on the right-hand side of Eq. (4.6). Moreover, in our approximation the intensity of the spontaneous sources [the right-hand part of Eq. (4.1)] may be assumed to be constant and independent of  $k\Omega$  and of time. Bearing these points in mind, we can rewrite Eq. (4.1) in the form

$$\frac{du_{\mathbf{k}}}{dt} - [(t - t_{\mathbf{th}})\dot{\boldsymbol{\beta}} - k^2\Omega^2 D] u_h = V, \qquad (4.7)$$

where the following notation is used:

$$D = \frac{\alpha}{(\Delta\omega \ \text{lum})^2} + \frac{\alpha 1}{(\Delta\omega \ \text{res})^2}, \quad V = \hbar \omega_0 \alpha \frac{N_2}{N_2 - (N_1 g_2/g_1)}.$$
(4.8)

This approach shows very clearly the two characteristic properties of the system considered: the nonstationary nature of the gain and its frequency dependence. Equation (4.7) shows also that the noncentral modes satisfy the threshold condition somewhat later than the mode with k = 0.

Equation (4.7) has an exact solution which can be expressed in terms of the error function  $\Phi(z)$ :

$$u_{k}(t) = V\left(\frac{\pi}{\dot{\beta}}\right)^{1/2} \exp\left\{\frac{\dot{\beta}}{2}(t-t_{th})^{2} + Dk^{2}\Omega^{2}(t-t_{th}) - \frac{D^{2}k^{4}\Omega^{4}}{2\dot{\beta}}\right\}$$
(4.9)  
 
$$\times \left\{1 + \Phi\left[\sqrt{\frac{\dot{\beta}}{2}}\left(t-t_{th}) - \frac{D\Omega^{2}k^{2}}{\dot{\beta}}\right)\right]\right\}.$$

At the moment of interest to us (t), the argument of the error function for the most effectively excited modes becomes much larger than unity. Moreover, we shall assume (this will be confirmed by the results of subsequent calculations) that the term  $D^2k^4\Omega^4/2\beta$  can be dropped from the argument of the exponential function. In this way, we find that the excitation energy of the k-th mode at a time t is given by the expression

$$u_{h}(t) = 2V \left(\frac{\pi}{\dot{\beta}}\right)^{1/2} \exp\left\{\frac{\dot{\beta}}{2} (t - t_{th})^{2} - k^{2}\Omega^{2}D (t - t_{th})\right\}.$$
 (4.10)

This expression shows that in the linear stage the dis-

tribution of the excitation energy over the modes is of Gaussian form:

$$u_{k}(t) \sim \exp\left\{-\frac{k^{2}\Omega^{2}}{[\Delta\omega(t)]^{2}}\right\}, \quad \Delta\omega(t) = \frac{1}{\sqrt{D(t-t_{\text{th}})}}, \quad (4.11)$$

where the effective width of the spectrum  $\Delta\omega(t)$  decreases with time proportionally to  $1/\sqrt{t-t_{th}}$ . The total energy, carried by all the modes, increases in accordance with the law

$$\sum_{k=-\infty}^{+\infty} u_k(t) \approx \int_{-\infty}^{+\infty} dk u_k(t) = \frac{2\pi V}{\Omega} \frac{\exp\{\dot{\beta} (t-t_{\rm th})^2/2\}}{V \,\dot{\beta} D (t-t_{\rm th})}; \qquad (4.12)$$

the dependence  $\exp{\{\dot{\beta}(t-t_{th})^2/2\}}$  is related to the non-stationary nature of the gain.

We shall now find the following quantities: 1) the width of the emission spectrum  $\Delta\omega(t_1)$  at a moment  $t_1$  corresponding to the onset of the nonlinear action of the filter; 2) the value of the gain  $\beta(t_1) = \alpha + (t_1 - t_{th})\dot{\beta}$  which is established at this moment. First, we find the moment of onset of the bleaching process  $t_1$  from the equation

$$I_{1} = \frac{c}{SL} \sum_{k=-\infty}^{+\infty} u_{k}(t_{1}); \qquad (4.13)$$

here, c is the velocity of light; L is the length of the resonator (in cm); S is the area of the cross section (in cm<sup>2</sup>) occupied by the modes with the lowest transverse indices; I<sub>1</sub> has the dimensions of erg  $\cdot$  cm<sup>-2</sup>  $\cdot$  sec<sup>-1</sup>. In the lowest approximation I<sub>1</sub> can be regarded as the power density at which significant bleaching of the filter takes place: I<sub>1</sub> ~ I<sub>b</sub>?. More accurately, we should substitute for I<sub>1</sub> a quantity considerably smaller than I<sub>b</sub>?. A detailed analysis of the bleaching process<sup>[9,10]</sup> shows that the effective interaction of light with a nonlinear filter in a laser begins when the instantaneous radiation power density reaches

$$I_{\text{inst}} \sim I_{\text{bl}} = \frac{\beta(t_1) - \alpha_1 - \alpha_{\text{nl}}}{\alpha_{\text{nl}}}$$
 (4.14)

(this point is discussed also in the next chapter, which deals with the bleaching process). Moreover, the fluctuation pattern of the radiation includes some peaks and the instantaneous power density  $I_{inst}$  for the strongest peaks may exceed the average value  $cL^{-1}S^{-1}\Sigma u_k$  by a factor of  $\ln[\Delta\omega(t)/\Omega]$  (see Chap. 6); these are the peaks of greatest interest to us. When these points are borne in mind, the equation for the determination of  $t_1$ , which is the moment from which the nonlinearity of the filter becomes significant, assumes the form

$$I_{1} = I_{bl}, \quad \frac{(t_{1} - t_{th})\beta}{\alpha n l} \frac{1}{\ln[1/\Omega \sqrt{D(t_{1} - t_{th})}]} = cS^{-1}L^{-1}\frac{2\pi}{\Omega} \hbar \omega_{0} \frac{N_{2}}{N_{2} - (N_{1}g_{2}/g_{1})} \alpha \frac{1}{\sqrt{D\beta(t_{1} - t_{th})}} \exp{\{\beta(t_{1} - t_{th})^{2}/2\}}.$$
(4.15)

We shall solve this equation by introducing the following dimensionless parameters:

$$Z = \beta (t_1 - t_{\text{th}})^2, \qquad (4.16a)$$

$$Y = \alpha^2 / \dot{\beta}, \qquad (4.16b)$$

$$m_0 = \frac{\Delta \omega \, \text{lum}}{\Omega} \left( 1 + \frac{\alpha_1}{\alpha_1 + \alpha_n l} \frac{(\Delta \omega \, \text{lum})^2}{(\Delta \omega \, \text{res})^2} \right)^{-1/2}$$
(4.16c)

$$A = \frac{I_{\rm bl} \quad SL}{2\pi\hbar c\omega_0} \frac{N_2 - (N_1 g_2/g_1)}{N_2}.$$
 (4.16d)

Equation (4.15) then becomes

$$\exp\left\{\frac{Z}{2}\right\} = Am_0^{-1}Y^{-3/4} \frac{Z^{3/4}}{\ln(m_0Y^{-1/4}) - \ln(Z^{1/4})}.$$
 (4.17)

The parameter m<sub>0</sub> represents the number of modes which can be fitted into the luminescence line and the root factor in Eq. (4.16c) gives the correction which allows for possible selective losses in the resonator. The parameter A represents the ratio of the bleaching power density Ibl to the power density corresponding to the optical energy of one photon  $\hbar\omega_0$  stored in the resonator. The quantity  $Am_0^{-1}$  is the ratio of the bleaching power density to the power density of the spontaneous radiation in the form of modes with the lowest transverse indices. The parameter Y represents the rise time of the gain, divided by the decay time of the field in an empty resonator. Finally, the parameter Z/2 determines the argument of the exponential function in the law describing the rise of the energy of the optical oscillations. The value of  $I_{bI}$  depends on the actual sub-stance used in the filter: by way of an estimate we can assume that  $I_{bI} \sim 2 \text{ MW/cm}^2 (\sec^{\lfloor 47,48 \rfloor})$ . We shall use the following typical values:  $S = 0.1 \text{ cm}^{-2}$ .  $L = 10^2 \text{ cm}$ ,  $\hbar\omega_0 \sim 10^{-19} \text{ J}$ ,  $\Delta\omega_{\text{lum}}/2\pi \text{ c} \sim 100 \text{ cm}^{-1}$ ,  $T_{\text{pump}} \sim 2 \times 10^4$ ,  $\times 10^{-3}$  sec. We then find that  $A \sim 10^{15}$ ,  $m_0 \sim 3 \times 10^4$ , and  $Y\,\sim\,5\times10^{5}.$  Since A is a very large number, we can easily find an approximate solution of Eq. (4.17):

$$Z = Z_1 + Z_2 + \dots,$$

$$Z_1 = 2 \ln \frac{A}{m_0 Y^{3/4} \ln (m_0 Y^{-1/4})}, \quad Z_2 = 2 \ln \frac{Z_1^{3/4}}{1 - [\ln (Z_1^{3/4})/\ln (m_0 Y^{-1/4})]}.$$
(4.18)

For these parameters, the value of Z is  $\sim 50$ . When the parameters of the resonator and the filter are varied, the value of Z is hardly affected because it is a logarithmic function of these parameters. We shall now express all the laser characteristics at the onset of the bleaching process  $(t_1)$  in terms of the quantities defined in the preceding paragraphs:

$$\beta(t_{i}) = \alpha \left[1 + \sqrt{Z/Y}\right], \qquad (4.19)$$

$$\Delta \omega(t_{i}) = m_{0} \Omega \left(YZ\right)^{-1/4} \equiv \frac{\Delta \omega_{lum}}{\sqrt{1 + \frac{\alpha_{1}}{\alpha_{1} + \alpha_{nl}} \frac{\Delta \omega_{lum}^{2}}{\Delta \omega_{res}^{2}}} \qquad (4.20)$$

In later chapters we shall need the parameters  $m = \Delta \omega(t_1)/\Omega$  and  $p = 1 + \alpha_n l/(\beta(t_1) - \alpha_l - \alpha_n l)$ . These parameters are given by

$$p = 1 + \frac{\alpha_{nli}}{\alpha_1 + \alpha_{nl}} \sqrt{\frac{Y}{Z}}, \qquad (4.21)$$

$$m = m_0 Y^{-1/4} Z^{-1/4}. \tag{4.22}$$

In the specific case we are considering we may assume that  $\alpha_{nl}/(\alpha_l + \alpha_{nl}) \sim 0.8$  and, therefore,  $p \sim 80$ ,  $m \sim 600$ . It would be interesting also to find the time from the moment of reaching the threshold to the onset of the nonlinear interaction with the filter:

$$t_{\rm i}-t_{\rm th}=\sqrt{Z}/\sqrt{\dot{\beta}}; \qquad (4.23)$$

in our example  $t_1 - t_{th}$  amounts to  $\sim 3 \times 10^{-5}$  sec. It is interesting to note that the product  $m\sqrt{p}$ , encountered in Chap. 5, is given by

$$m \sqrt{p} = \sqrt{\frac{\alpha_{\rm q_l}}{\alpha_1 + \alpha_{\rm nl}}} m_{\rm q} \frac{1}{\sqrt{Z}}, \qquad (4.24)$$

All the expressions obtained so far contain the parameter

$$\dot{\beta} \equiv \frac{d\beta}{dt}\Big|_{t=t}$$
 th

The value of this parameter depends on the actual nature of the function  $\beta(t)$  and has to be determined separately for each laser system. By way of illustration, we shall derive the expression for  $\beta$  for a simple pumping model. Let us assume that

$$\beta(t) = \beta_{\max} \left[ 1 - \left( \frac{t}{T_{pump}} \right)^2 \right], \qquad (4.25)$$

where the gain  $\beta_{\text{max}}$  can be expressed in terms of the absorption coefficient  $\alpha = \alpha_l + \alpha_{nl}$  and the excess of the pumping energy over the threshold  $\eta$ :

$$\beta_{\max} = \alpha \frac{W_{pump}}{W_{pump, th}} \equiv \alpha \eta.$$
 (4.26)

In this case, the quantity  $\dot{\beta}$  is given by

$$\dot{\beta} = \frac{2\alpha}{T_{\text{pump}}} \sqrt{\eta (\eta - 1)}.$$
(4.27)

The reader may wish to substitute the above expression into the formulas for p and m.

Thus, we have been able to express the quantities p and m in terms of laser parameters which can be determined by direct experiments:

$$\alpha_i$$
,  $\alpha_{ni}$ ,  $I_{bi}$ ,  $\Delta \omega_{lum}$ ,  $\Delta \omega_{res}$ ,  $\eta$ ,  $T_{pump}$ .

#### 5. TRANSFORMATION OF THE FIELD PROFILE IN THE FILTER BLEACHING PROCESS

We have shown earlier that a quasiperiodic fluctuation profile of the field  $\mathcal{E}_0(t)$  is established before the onset of the bleaching process. We shall now consider the transformation of this profile during the interaction of the field with the filter.

We must first formulate more exactly what we understand by the initial and final profiles of the field. Estimates show that the duration of the bleaching process in real systems is relatively short:  $\sim 5 \times 10^{-7}$  sec. In view of this, we can consider the bleaching process ignoring the nonstationary nature of the gain and its frequency dependence (these two effects are most important in the preceding linear stage).

Bearing these points in mind, we can describe the field in the laser before the bleaching process by

$$E_{\text{before bleach}}(t) = e^{\frac{1}{2}(\beta - \alpha_1 - \alpha_{nl})t} \mathscr{E}_0(t), \qquad (5.1)$$

where  $\mathscr{E}_0(t)$  is a periodic function of time:

$$\mathscr{E}_{0}\left(t+T\right) = \mathscr{E}_{0}\left(t\right) \tag{5.2}$$

(T is the period of the laser resonator). This periodic function  $\mathcal{S}_0(t)$  will be called the initial field profile or the profile before bleaching.

After the end of the bleaching process, i.e., when the filter becomes transparent even at the lowest point of the field profile, the rise of the field in the laser is exponential and the filter losses do not occur in the argument of the exponential function:

$$E_{\text{after bleach }}(t) = e^{\frac{1}{2}(\beta - \alpha_{1})t} \mathscr{E}(t).$$
(5.3)

The periodic function  $\mathscr{E}(t)$ ,

$$\mathscr{E}(t) = \mathscr{E}(t+T), \tag{5.4}$$

will be called the field profile after bleaching.

Since the bleaching stage is relatively short, the

values of the gain  $\beta$  which occur in Eqs. (5.1) and (5.3) can be assumed to be identical and equal to the gain at the center of the line which is established before bleaching.

Thus the problem reduces to the derivation of the relationship between the initial field profile  $\mathcal{S}_0(t)$  and the function  $\mathcal{S}(t)$ , which is the profile after bleaching. This problem has been solved in<sup>[9,10]</sup>.

We shall now make some more definite assumptions relating to the laser system itself. We shall consider a traveling-wave laser (for example, a unidirectional ring laser) which contains a bleachable filter. The unidirectional nature of the filter simplifies theoretical analysis because it allows us to ignore the possibility of simultaneous bleaching of the filter by pulses traveling in opposite directions. The same is also true of a standing-wave laser if the cuvette containing the bleachable material is sufficiently thin:  $l \stackrel{<}{\scriptstyle \sim} c au_n$ , and if this cuvette is pressed tightly against one of the resonator mirrors. Moreover, the bleachable filter should have instantaneous response (it should respond to instantaneous changes in the field intensity) in order to provide the most favorable conditions for the formation of ultrashort light pulses. Therefore, we shall assume that the filter has an instantaneous response. Finally, we shall postulate that before bleaching the field rises slowly:  $(\beta - \alpha_l - \alpha_{nl})T/2 \ll 1$ . Estimates made in the preceding chapter show that this inequality is satisfied at practically all the values of the pumping level because at the onset of bleaching the gain is always very close to its threshold value. In fact,  $(\beta - \alpha_l - \alpha_{nl})T = p^{-1}(\beta - \alpha_l)T$ , where p is much larger than unity, and if the gain after bleaching  $\exp\{(\beta - \alpha_l)T/2\}$  is not too high  $[(\beta - \alpha_l)T/2 \lesssim 1]$ , this inequality is satisfied. When these assumptions are made, the transformation of the field profile can be found in its general form.

We shall select two points in the initial profile in such a way that they satisfy

$$|\mathscr{E}_{0}(t_{1})| = |\mathscr{E}_{0}(t_{2})| e^{(\beta - \alpha_{1} - \alpha_{n_{1}})Tq/2}, \qquad (5.5)$$

where q is an integer. In this case, the evolution of the field at a point  $t_2$  in one period during the filter bleaching process is given by the same law as at the point  $t_1$  but with a lag of q periods. After bleaching, the shift by q periods gives rise to a change in the amplitude

amounting to a factor of  $\exp\{(\beta - \alpha_l) \operatorname{Tq}/2\}$ . Therefore, in the final profile we have

$$|\mathscr{E}(t_1)| = |\mathscr{E}(t_2)| e^{(\beta - \alpha_1)Tq/2}.$$
(5.6)

Combining Eqs. (5.5) and (5.6), we find that

$$\frac{|\mathfrak{C}(t_2)|}{|\mathfrak{C}(t_1)|} = \left[\frac{|\mathfrak{C}_0(t_2)|}{|\mathfrak{C}_0(t_1)|}\right]^{\beta - \alpha_1 - \alpha_{n_1}}.$$
(5.7)

The transformation law given by Eq. (5.7) applies to all points which satisfy the condition (5.5) with an integral value of q. Since the dependence of  $\mathscr{E}$  on  $\mathscr{E}_0$  is monotonic and since the network of points with integral values of q is sufficiently "fine" [because  $\exp\{(\beta - \alpha_l - \alpha_{nl})T/2\} - 1 \ll 1]$ , it follows that Eq. (5.7) describes satisfactorily the transformation of the whole initial function  $\mathscr{E}_0(t)$ . Therefore, the transformation of the amplitude profile of the field can be written in the form

$$|\mathcal{E}(t)| = \operatorname{const.} |\mathcal{E}_0(t)|^p, \qquad (5.8)$$

$$p = \frac{\beta - \alpha_l}{\beta - \alpha_l - \alpha_{nl}} . \tag{5.9}$$

We shall assume that—in the laser system considered—the phase of the field is not transformed, i.e., that

$$\frac{\mathscr{E}(t)}{|\mathscr{E}(t)|} = \frac{\mathscr{E}_{\cap}(t)}{|\mathscr{E}_{\cap}(t)|} \,. \tag{5.10}$$

Thus, the law of transformation of the field profile during the bleaching of a filter in a laser is of the form<sup>4</sup>)

$$\mathscr{E}(t) = \operatorname{const} \cdot \frac{\mathscr{E}_0(t)}{|\mathscr{E}_0(t)|} | \mathscr{E}_0(t)|^p.$$
(5.11)

The constant in Eq. (5.11) represents the duration of the bleaching process and it cannot be determined accurately in this very general form. This constant is calculated in  $[^{9,10}]$  for a specific laser system and a definite model of the bleaching process. However, we shall not be interested in the exact value of this constant. The most important characteristic of the transformation law is the nonlinearity exponent p defined by Eq. (5.9). The value of the laser in the linear stage and has been calculated in the preceding chapter.

The true measure of the deformation of the initial function is the quantity  $p - 1 = \alpha_{n l}/(\beta - \alpha_l - \alpha_{n l})$  because the transformation of Eq. (5.11) is linear if p = 1. The meaning of the parameter p - 1 can be found from the following simple estimates. The order of magnitude of the duration of the bleaching process is

$$\Delta t_{\rm bl} \approx (\beta - \alpha_l - \alpha_{\rm nl})^{-1} \qquad (5.12)$$

or, in terms of the number of field passes through the filter,

$$Q_{\rm bl} \approx \Delta t_{\rm bl} / T \approx [T (\beta - \alpha_{\rm l} - \alpha_{\rm nl})]^{-1}.$$
 (5.13)

During this time interval the filter acts essentially as a nonlinear element. A characteristic of the effect of the filter per single pass is its absorption coefficient  $\alpha_{nl}T$ . A measure of the total effect of the nonlinear filter is the product

$$Q_{bl} \cdot \alpha_{nl} T = \frac{\alpha_{nl}}{\beta - \alpha_l - \alpha_{nl}} \cdot$$
 (5.14)

Hence, we can easily see why this quantity occurs in the field transformation law. The nonlinearity exponent may be increased by increasing the optical density of the filter  $\alpha_{nl}$  (enhancing the effect of the filter per single pass) or by increasing the number of effective passes  $Q_{bl}$ , which means that the laser should operate near the threshold:  $\beta - \alpha_l - \alpha_{nl} \rightarrow 0$ .

It follows from Eq. (5.11) that the deformation of the initial field profile (and, therefore, of the intensity profile  $I(t) = |E(t)|^2$ ) is manifested in two aspects. Both

these aspects were pointed out qualitatively by DeMaria, Stetser, and Heynau.<sup>[1]</sup>

First, each intensity peak of the initial profile becomes narrower by a factor of  $\sqrt{p}$  on the time scale. We shall illustrate this for a Gaussian pulse:

$$I_0(t) = Ae^{-(t-t_0)^2/\tau_1^2}, \quad I(t) = \text{const} \cdot [I_0(t)]^p = \text{const}' \cdot e^{-p(t-t_0)^2/\tau_1^2}.$$
 (5.15)

The reduction in the pulse duration corresponds to the broadening of the field spectrum by a factor of  $\sqrt{p}$ . The final pulse duration becomes [compare with Eq. (4.24)]

$$\mathbf{r}_{\text{final}} = \frac{\tau_1}{\sqrt{p}} = \frac{1}{\Delta \omega_1 \sqrt{p}} = \tau_0 \sqrt{\frac{\alpha_{\text{nl}} + \alpha_1}{\alpha_{\text{nl}}}} \sqrt{\overline{z}}, \qquad (5.16)$$

where  $\tau_0 = \Delta \omega_{lum}^{-1} [1 + \alpha_l \Delta \omega_{lum}^2 / (\alpha_l + \alpha_n l) \Delta \omega_{res}^2]^{1/2}$  is the peak duration corresponding to the width of the luminescence line (with a correction for the selective losses in the resonator). Under realistic conditions  $\sqrt{Z}$ ~ 6-8 and Eq. (5.16) yields an almost universal expression for  $\tau_{final}$ ; this means that the narrowing of the spectrum in the linear stage is almost completely compensated (to within a factor of 7) by the subsequent broadening of the spectrum during the stage of bleaching of the instantaneous-response filter.

Secondly, if any two peaks in the initial field profile have amplitudes  $I'_0$  and  $I''_0$ , respectively, the ratio of the intensities of the corresponding peaks in the profile obtained after bleaching is given by

$$\frac{l'}{l''} = \left(\frac{l'_0}{l'_0}\right)^p.$$
 (5.17)

Since  $p \gg 1$ , Eq. (5.17) implies that the inequality of two peaks in the initial profile becomes much greater after the bleaching stage. In particular, the most desirable situation is that when the final intensity profile is dominated by a single pulse. The probability of achieving this situation is calculated in the next chapter (see Chap. 6) as a function of the laser parameters.

We have considered the time dependence of the periodic function  $\mathscr{E}(t)$ . This time dependence is particularly convenient in the analysis of the field transformation by the bleachable filter. On the other hand, the periodicity of the function  $\mathscr{E}(t)$  makes it possible to describe the behavior of the field in terms of discrete Fourier components which are practically identical (in our case) with the amplitudes of modes having different longitudinal indices:

$$\mathscr{E}(t) = e^{-i\omega_0 t} \sum_{h=-\infty}^{+\infty} e^{-ih\Omega t} \mathscr{E}_h(t).$$
(5.18)

This approach has been employed in the first investigations of laser with bleachable filters and the first explanations have been based on this approach ( $\sec^{[2]}$ ). The spectral or mode approach may be more convenient in the case when only one radiation pulse is emitted per period. In this case, the phases of all the modes depend linearly on the mode number:

$$\mathcal{E}_{k} = |\mathcal{E}_{k}| e^{+ik\Omega t_{0}}, \qquad (5.19)$$

and the field itself will be in the form of a single pulse per period with its maximum located at the point  $t_0$  and its width given by  $\Delta t_p \sim 1/\Delta \omega$ :

$$\mathscr{E}(t) = f(t - t_0), \quad f(\tau) = \sum |\mathscr{E}_h| e^{-ik\Omega\tau}. \quad (5.20)$$

In view of this, the formation of a train of ultrashort

<sup>&</sup>lt;sup>4)</sup>Strictly speaking, the field profile described by Eq. (5.11) can be established only after a time sufficient for even the lowest points of the profile to grow in amplitude so that the filter becomes transparent at these points. In real lasers the saturation of the amplifying medium limits the total duration of the stimulated emission process, including the amplifying stage. Therefore, the transformation law of Eq. (5.11)does not apply to the lowest points of the field profile. Nevertheless, since only the strongest peaks in the final profile are of importance in the problems of interest to us, the use of the asymptotic transformation law (5.11) does not lead to significant errors.

pulses is frequently called the mode self-locking.

However, the spectral or mode approach is quite unsuitable if we wish to find whether a single strong pulse will evolve from the initial fluctuation pattern. In fact, when the initial fluctuation profile is transformed in accordance with Eq. (5.11), the field spectrum broadens by a factor of  $\sqrt{p}$ . Each initial fluctuation peak with its maximum at the point  $t = t_j$  makes a contribution to the amplitude of the k-th mode and this contribution is proportional to  $\exp(ik\Omega t_j)$ . The initial fluctuation pattern includes a large number of irregularly distributed peaks. Therefore, the complex amplitude of the k-th mode is the sum of the contributions of each of the pulses:

$$\mathscr{E}_{k} \sim \sum_{j} b_{j} e^{i k \Omega t_{j}}; \qquad (5.21)$$

in general, the phase of  $\mathcal{Z}_k$  is a complex nonmonotonic function of the mode number k.

Let us consider again the transformation of the field during the bleaching stage. The two effects-the narrowing of the pulses and the selection of the strongest pulses-occur simultaneously in a single process of interaction between the field and the filter. However, only the second effect is equivalent to the phasing of the modes in the initial spectrum. It is this effect that gives rise to a regular time distribution which evolves from the initial random time sequence. The narrowing of the pulses represents-in the spectroscopic language-the appearance of new spectral components which is not related to the phasing of the initial components. We can easily imagine a laser model in which mode phasing is not accompanied by broadening of the spectrum. For example, the dispersion of the gain may prevent such broadening. If the initial field in a laser with a bleachable filter is of spectral width which identical with the total width of the gain profile, it is found that the broadening of the spectrum is unlikely but the phasing effect remains. This effect is again manifested by a preferential growth of the strongest pulses.

## 6. STATISTICS OF THE APPEARANCE OF ULTRASHORT PULSES DURING FILTER BLEACHING

We have mentioned earlier that the formation of single ultrashort light pulses in a laser with a bleachable filter is due to the selection of such pulses from the initial fluctuation pattern which forms in the laser before the onset of the bleaching process.

The transformation of the fluctuation pattern during the bleaching of a filter was first calculated by  $Fleck^{[57]}$ on a computer. Unfortunately, it was not possible to carry out a similar analysis by the method employed  $in^{[57]}$ :  $Fleck^{[57]}$  computed the only realization of a random process resulting from the presence of just 11 initial modes. Therefore, we shall use an analytic expression (5.15) obtained in Chap. 5 for the transformation of the field profile during the bleaching of an instantaneousresponse filter. In the present chapter, we shall calculate, following the work of Kuznetsova,<sup>[13]</sup> the probability of the appearance of single light pulses in one resonator period as a result of transformation of the initial fluctuation pattern during the bleaching of the filter. We shall consider first the statistical properties of the initial field profile and concentrate our attention on the presence of strong peaks. It is essential to know the statistical properties of the initial profile if we wish to answer the question of whether the profile transformed by the action of the filter consists of just one strong pulse per period. In view of the statistical nature of the process, we must calculate the probability of this event.

An important problem in the statistics of peaks in the initial profile was formulated by Letokhov.<sup>[55,56]</sup> However, Letokhov's treatment cannot be used directly in the solution of our problem. We shall follow the work of Kuznetsova.<sup>[13]</sup>

The initial field profile is formed as a result of linear amplification of noise fields which are generated as a result of the spontaneous emission of radiation by atoms in the resonator. Since the amplification is linear and the number of statistically independent spontaneous noise sources (atoms) is large, the initial field profile  $\mathscr{E}_0(t)$  (the profile before bleaching) can be regarded as a complex Gaussian random process.<sup>5)</sup>

The probability distribution of the field modulus  $|\mathscr{F}_0(t)|$  in the case of a complex Gaussian random process is given by the well-known Rayleigh law. It is convenient to express this law in terms of the field intensity:  $I_0(t) = |\mathscr{F}_0(t)|^2$ . The distribution of  $I_0$  is then of the form

$$v(I_0) dI_0 = \frac{1}{\langle I_0 \rangle} \exp \left\{ -\frac{I_0}{\langle I_0 \rangle} \right\} dI_0.$$
 (6.1)

The mean-square fluctuation of this quantity is

$$\langle \Delta I_0^2 \rangle = \langle I_0 \rangle^2, \tag{6.2}$$

and the correlation of the intensity is given by the expression

$$\langle (I_0(t) - \langle I_0 \rangle) (I_0(t+\tau) - \langle I_0 \rangle) \rangle = \langle I_0 \rangle^2 | \gamma_0(\tau) |^2;$$
(6.3)

here,  $\gamma_0(\tau)$  is the normalized correlation function of the field  $\mathscr{E}_0(t)$  determined by its spectral power density:

$$\gamma_0(\tau) = \frac{\langle E_0^*(t) E_0(t+\tau) \rangle}{\langle |E_0|^2 \rangle} = \int_{-\infty}^{+\infty} g(\omega) e^{-i\omega\tau} d\omega.$$
 (6.4)

The value of  $|\gamma_0(\tau)|$  during a time interval  $|\tau| \leq T$  differs significantly from zero only when  $|\tau| \leq (\Delta \omega_1)^{-1}$ , where  $\Delta \omega_1$  is the width of the spectrum established at the beginning of the bleaching process.

It follows from Eqs. (6.1)–(6.4) that the intensity  $I_0(t)$  fluctuates about a mean value  $\langle I_0 \rangle$  by an amount which is of the order of  $\langle I_0 \rangle$  and the intensity maxima change to minima in a time  $\sim \tau_{\rm corr} = (\Delta \omega_1)^{-1}$ . The number of the intensity maxima (or minima) per unit time is of the order of  $\tau_{\rm corr}^{-1} = \Delta \omega_1$ . We must stress that the function  $\mathscr{F}_0(t)$  is periodic, i.e., we are dealing with a periodic random process characterized by a period T. In realizations of the random process of duration T the number of maxima fluctuates from one realization to another and the average number of maxima is of the order of

$$m \approx T/\tau_{\rm corr} = T \Delta \omega_1/2\pi.$$
 (6.5)

<sup>&</sup>lt;sup>5)</sup>To avoid misunderstanding we must stress that we are speaking of the Gaussian nature of the *statistics* of the process, i.e., we shall be speaking of the probability distributions, etc. The fact that the spectrum of the statistically Gaussian process is of the form  $g(\omega) \propto \exp \left\{-(\omega - \omega_0)^2/\Delta \omega_1^2\right\}$ , i.e., it is also described by the Gaussian function, it is not related directly to the statistical problems that we shall now consider.

Since  $T = 2\pi/\Omega$ , the parameter m is of the same order of magnitude as the number of the modes which are excited effectively in the initial spectrum.<sup>6)</sup> The expression for the parameter m, expressed in terms of the laser characteristics, has been derived in Chap. 4.

Unfortunately, there are very few exact formulas suitable for the determination of the number of maxima and the distribution of their amplitudes in a Gaussian random process of finite duration (see the work of Tikhonov<sup>[14,15]</sup>). We must stress that our problem is even more complex because we are interested no so much in the initial Gaussian process but principally in the result of its transformation by the action of a bleachable filter, i.e., we are interested in the p-th order of a Gaussian process with  $p \gg 1$ . Therefore, instead of the initial Gaussian process  $\mathscr{F}_0(t)$ , we shall consider a model random process  $\mathscr{F}_0(t)$  and, at the same time, allows us to calculate the statistical characteristics of interest to us.

We shall start with a random function in the form of a train of a fixed number (m) of nonoverlapping pulses:

$$\mathscr{E}_{1}(t) = C \sum_{n=1}^{m} A_{n} f(t - \tau - t_{n});$$
 (6.6)

here, f(t) is a pulse function of period T whose value differs from zero only in the interval  $0 \le t \le T/m$ . The actual form of the pulse function is of no significance; however, it is important that the pulses should not overlap. We shall assume that the pulses are equidistant:  $t_n = (T/m)n$ , where n is the number of the pulse. We shall use Eq. (6.5) to select the number of pulses m in accordance with the spectral width of the real Gaussian process  $\mathcal{F}_0(t)$ . We shall assume that the shift  $\tau$  is distributed uniformly in the interval  $0 \le \tau \le T/m$  and that the complex amplitudes of the pulses are Gaussian random quantities independent of one another.

We shall introduce the phase and amplitude of the pulses:

$$A_n = \sqrt{x_n} e^{i\varphi_n}. \tag{6.7}$$

The phases of the pulses are assumed to be distributed uniformly in the interval  $(0, 2\pi)$  and the intensities have a distribution of the type

$$v(x_n) dx_n = \exp\{-x_n\} dx_n.$$
 (6.8)

The normalization constant C in Eq. (6.6) will be selected, for the sake of convenience, so as to give  $\langle x_n \rangle = 1$ .

We shall use  $n_1$  to denote the pulse whose intensity is greater than the intensities of all the other pulses in a given period. The probability distribution for  $x_{n_1}$  is of the form

$$\widetilde{w}(x) \, dx = m e^{-x} \left[ 1 - e^{-x} \right]^{m-1} dx. \tag{6.9}$$

The quantity  $\widetilde{w}(x)$  has a maximum at the point  $x = \ln m$ . This means that the most probable value of the intensity of the strongest pulse is several times higher than the average intensity if the number of pulses m in a period is sufficiently large.

An important characteristic in our problem is not

the absolute value of the amplitude of the strongest peak (this has been estimated  $in^{[55,56]}$ ) but the amplitude of this peak relative to the amplitudes of other peaks in the same realization.

If the value of m is high, other pulses of amplitude close to that of the strongest pulse are quite likely to appear. The distribution of the probability for the second strongest pulse  $x_{n_0}$  is of the form

$$\widetilde{w}(x) \, dx = m \, (m-1) \, e^{-2x} \, [1 - e^{-x}]^{m-2} dx \tag{6.10}$$

and the maximum of this probability occurs at  $x = \ln (m/2)$ .

The relationship between the strongest and all the other pulses can be obtained from the probability that the amplitude of the strongest pulse exceeds the amplitudes of all the other pulses by a factor which is not less than a:

$$W\{x_{n_1} \ge ax_n\} = \frac{\Gamma(a+1)\Gamma(m+1)}{\Gamma(a+m)},$$
 (6.11)

where  $\Gamma(z)$  is the gamma function or the Euler integral of the second kind. This formula will be needed later and can be derived as follows. The probability  $W\{x_{n_1} \geq ax_n\}$  consists of m identical terms. The first term represents the probability that the first pulse is the strongest and the inequality  $x_1 \geq ax_n$ ,  $n \neq 1$  is satisfied. The second term represents the probability that the second pulse is the strongest, etc.:

$$W = mW^{(n_1=1)}$$
 (6.12)

The probability  $W^{(n_1=1)}$  is given by the integral

$$W^{(n_1=1)} = \int_0^\infty dx \, w(x) \left[ P\left(\frac{x}{a}\right) \right]^{m-1}; \tag{6.13}$$

here, w(x) is the distribution of the probability of the intensity of the first pulse;  $P(\xi)$  is the probability that a particular pulse has an intensity  $x_n \leq \xi$ 

$$P(\xi) = \int_{0}^{\xi} w(x) \, dx, \qquad (6.14)$$

and the factor  $[P(x/a)]^{m-1}$  gives the probability that m-1 remaining pulses are at least a times weaker than the first pulse. Substitution of the expression  $w(x) = e^{-x}$  leads to

$$W\{x_{n_{1}} \geq x_{n_{1}} = m \int_{0}^{\infty} dx e^{-x} [1 - e^{-x/a}]^{m-1}$$

and this yields Eq. (6.11).

We shall now estimate the probability that the amplitude of the strongest pulse is twice as large as the amplitude of any of the other pulses. It follows from Eq. (6.11) that this probability is 2/(m + 1), i.e., it is very small if the value of m is large.

Of greatest interest is the situation in which one of the pulses in the final profile contains most of the energy emitted in one period. We shall denote the pulse amplitudes in the final profile by  $I_n$  (n = 1, 2, ..., m):

$$I_n = (x_n)^p$$
. (6.15)

We shall be interested in events such that the energy in the strongest pulse is at least M times as large as the energy in all the other pulses:

$$I_{n_1} \gg M \sum_{n \neq n_1} I_{n_1}$$
(6.16)

<sup>&</sup>lt;sup>6)</sup>Factors such as  $2\pi$ , etc., are unimportant in the determination of the parameter m because the expressions which we shall obtain depend logarithmically on m.

We shall call this situation the "pulse selection to a precision determined by the parameter M." The pulse selection process can also be regarded as the achievement of full mode self-locking. In view of the statistical nature of the process in question, we can speak only of the probability of pulse selection (the probability of full self-locking).

A direct analytic calculation of the probability of self-locking in the model considered, i.e., the calculation of the probability that the inequality (6.16) is obeyed, is very difficult. Therefore, we shall give the upper and lower limits of the probability that this will happen. We shall show that the probability W lies within the limits

$$F_2(p, m, M) < W < F_1(p, m, M),$$
 (6.17)

where the functions  $F_2(p, m, M)$  and  $F_1(p, m, M)$  are given by the expressions

$$F_2(p, m, M) = \frac{\Gamma(a_2+1)\Gamma(m+1)}{\Gamma(a_2+m)}, \quad a_2 = |M(m-1)|^{1/p}, \quad (6.18)$$

$$F_1(p, m, M) = \frac{\Gamma(a_1+1)\Gamma(m+1)}{\Gamma(a_1+m)}, \quad a_1 = M^{1/p}.$$
 (6.19)

The functions  $F_2$  and  $F_1$  represent the probabilities of events, the first of which is the special case of the event of interest to us and defined by Eq. (6.16) and the second is more general and includes the event of Eq. (6.16) as the special case. The function  $F_2$  represents the probability that the amplitude of the strongest pulse exceeds the amplitudes of all the other pulses by a factor of at least M(m-1):

$$I_{n_1} \ge (m-1) M I_n, \quad n \ne n_1;$$
 (6.20)

if the inequality (6.20) is satisfied, the inequality of Eq. (6.16) is also satisfied but the converse is not true. Consequently,  $W(6.20) = F_2 < W(6.16)$ .<sup>7)</sup> The expression (6.18) for the probability  $F_2$  is obtained by applying the inequality (6.20) to the initial profile,

$$x_{n_1} \ge [M(m-1)]^{1/p} x_n, \quad n \ne n_1,$$
 (6.21)

and using Eq. (6.11). The function  $F_1$  represents the probability that the amplitude of the strongest pulse in the final profile exceeds the amplitudes of all the other pulses by a factor of at least M:

$$I_{n_1} \gg M I_n, \quad n \neq n_1. \tag{6.22}$$

If the inequality (6.16) is satisfied, we know that the inequality (6.22) is also satisfied but the converse is not true and, therefore,  $W(6.22) = F_1 > W(6.16)$ .

The events represented by the inequality (6.22) with a given value of M may sometimes be of experimental interest and, therefore, Eq. (6.19), representing the probability of these events, is of special interest.

If the number of modes in the initial profile is large (m  $\gg$  1), Eq. (6.17) simplifies to

$$e^{-\frac{1}{p}[\ln m + \ln M]\ln m} < W < e^{-\frac{1}{p}\ln M \ln m}$$
 (6.23)

The inequalities (6.17) and (6.23) solve the problem of estimating the probability of full self-locking of modes. In the case of the laser considered in Chap. 4, we have p = 80 and m = 600. If we assume that M = 9, it follows from Eq. (6.23) that  $0.50 < W_{M=9} < 0.84$ . This means that, although the probability of mode self-locking is high (exceeding 50%), the inequality (6.16) does not apply to a significant number (exceeding 16%) of the laser pulses if M = 9.

The inequalities in Eq. (6.23) can be rewritten in a different manner. If we assume a particular value of the probability W(6.16) =  $\eta$  of mode self-locking (for example,  $\eta = 0.5$ ) and a specified value of the parameter M, we can show that the probability of self-locking (for this value of M) in the range

$$p \ge p_2(m, M, \eta) \approx \frac{(\ln m + \ln M) \ln m}{\ln (1/\eta)}$$
 (6.24)

is high:  $W > \eta$ , whereas the probability of self-locking (for the same parameter M) in the range

$$p \leqslant p_1(m, M, \eta) \approx \frac{\ln M \ln m}{\ln 1/\eta}$$
(6.25)

is low:  $W < \eta$ .

The curve in the (p, m) plane for which  $W(6.16) = \eta$ divides the regions with high and low probabilities of self-locking and it passes between the curves  $p_1(m)$  and  $p_2(m)$  corresponding to fixed values of M and  $\eta$ . We determined the exact position of this curve by numerical calculations on a computer in which a random number program was used (we are not aware of an analytic solution of this problem).

The relevant dependences are plotted in Fig. 2 for  $\eta = 0.5$  and M = 9. The lowest curve shows the dependence  $p_1(m)$  and the highest represents  $p_2(m)$ : these dependences were obtained using the analytic expressions in Eqs. (6.24) and (6.25). The middle curve (3) is the plot of the results of our numerical calculations: it represents the boundary of the region of full self-locking in the (p, m) plane for M = 9 and  $\eta = 0.5$ .

Similar curves are plotted in Fig. 3 for  $\eta = 0.5$  and M = 1.5. In the first case (Fig. 2, M = 9, a pulse contains at least 90% of the energy evolved in a period), the boundary of the full self-locking region lies closer to the dependence  $p_1(m)$ , whereas in the second case (Fig. 3, M = 1.5, a pulse contains at least 60% of the energy evolved in one period), the same boundary lies much further from  $p_1(m)$ .

It is clear from Figs. 2 and 3 that the larger the initial number m of the oscillation modes, the more difficult they are to lock. The curves plotted in these figures show that full self-locking of a large number of modes



FIG. 2. Dependence of the nonlinearity exponent p on the number of modes m established before the onset of bleaching;  $\eta = 0.5$ , M = 9. Above curve 3 (obtained by calculation) the probability that the strongest post-bleaching pulse represents at least 90% of the energy emitted per period is  $W > \eta = 0.5$  and below this curve the probability is  $W < \eta =$ 0.5.

<sup>&</sup>lt;sup>7)</sup>W(6.20) denotes the probability of the event or physical situation represented by Eq. (6.20).



FIG. 3. Same dependence as in Fig. 2 but for  $\eta = 0.5$ , M - 1.5 (it is assumed that the probability that the pulse contains at least 60% of the energy emitted in one period is  $W > \eta$ ). Below curve 1 the probability that the strongest pulse exceeds the next pulse by a factor of M = 1.5 is quite low:  $W_1 < \eta = 0.5$ . Above curve 2 the probability that the strongest pulse exceeds any of the other pulses by a factor of M(m-1) times is  $W_2 < \eta = 0.5$ .

m requires high values of the nonlinear conversion exponent p. This dependence of p on m is due to the fact that the probability of the appearance of several highintensity peaks of comparable amplitudes in the initial field is higher when the number of modes is large. Therefore, in this case, the nonlinearity must be of higher order p in order to achieve selection of the strongest pulse. This dependence of the probability of the appearance of a single pulse in one period on the width of the initial spectrum has been confirmed experimentally:<sup>[28]</sup> in this investigation the initial width of the spectrum was varied by introducing linear losses into the resonator of a laser with a sharp minimum at the center of the emission line. The time dependence of the intensity of the output radiation was recorded with a resolution of ~10<sup>-f1</sup> sec.

## 7. INFLUENCE OF GAIN SATURATION ON THE FINAL FIELD PROFILE

When the filter becomes completely transparent the intensity of the laser oscillations rises rapidly [the rise is proportional to  $\exp\{(\beta - \alpha_l)t\}$ ] in a relatively short time interval. This rise soon stops because the radiation extracts a considerable part of the energy stored in the active medium and this reduces the gain. Beginning from some particular moment (corresponding to the peak of a giant pulse), the gain falls below the value of the losses and the field begins to decay.

We are interested in producing a regular train of very short light pulses and, therefore, we have to ask whether the saturation of the laser transition can impair the intensity profile which is formed under the influence of the bleachable filter. In other words, we have to enquire whether the inversion saturation may lead to impairment of the relationship between the strongest pulse and the rest of the radiation in the vicinity of the giant pulse maximum.

We shall answer these questions by finding the evoluation of the intensity profile of the laser radiation, making allowance for the inversion depletion in the active substance. We shall show that, in cases of practical interest, the saturation of the active material does not give rise to a significant distortion of the time dependence of the radiation in one period.

We shall consider once again a ring laser. We shall assume that all the elements of this laser, with the exception of the active substance, are linear in the stage being considered here. The propagation of the radiation in the amplifying medium has often been discussed in the literature<sup>[16,17,49]</sup></sup> making allowance for the saturation of the active transition. The equations for theintensity I and inversion N are of the form</sup>

$$\frac{\partial I}{\partial z} + \frac{1}{v} \frac{\partial I}{\partial t} = N \sigma I, \qquad (7.1)$$

$$\frac{\partial R}{\partial t} = -2N\sigma I, \qquad (7.2)$$

where  $\sigma$  is the cross section of the laser transition. We shall assume that the process of formation of a giant pulse after the bleaching stage is quite rapid. Therefore, the influence of pumping and of population relaxation can be dropped from the equation for  $\partial N/\partial t$ . The solution of both equations is

$$I(z, t) = \frac{I(z=0, t-\frac{z}{v})}{1-(1-e^{-z\sigma N_0})\exp\left[-\sigma\int_{-\infty}^{t-(z/v)}I(z=0, t')dt'\right]},$$
 (7.3)

where N<sub>0</sub> is the initial inversion. In one period in a ring laser the radiation traverses an active element of length l, is reflected from mirrors characterized by a general loss factor  $\exp(-\alpha_l T)$ , and approaches the active medium again. Therefore, at any point in the resonator the following equation is satisfied:

$$I(t+T) = e^{-\alpha_{I}T}I(t) - \frac{1}{1 - (1 - e^{-\beta_{T}}e)e^{-\alpha_{T}}};$$
(7.4)

here,  $\exp(\beta T) \equiv \exp(N_0 \sigma l)$  is the unsaturated gain of the active medium per period. We shall introduce the functions

$$r(t) = \sigma \int_{-\infty}^{t} I(t') dt', \quad I(t) = \frac{1}{\sigma} \frac{dr}{dt}.$$
 (7.5)

Integrating Eq. (7.4), we obtain

$$r(t+T) = e^{-\alpha} i^{T} \ln \left[ (e^{r(t)} - 1 + e^{-\beta T}) e^{+\beta T} \right].$$
 (7.6)

This equation was first derived by Gurevich.<sup>[18]</sup> A characteristic feature of Eq. (7.6) is that it relates the values of the function r(t) at discrete points separated from one another by the resonator period T. It is this that allows us (as in Chap. 5) to draw definite conclusions about the relationship between the final and initial field profiles.

Equation (7.6) has two singular solutions such that r(t) = r(t + T):

$$r_{i,2} = e^{-\alpha} l^T \ln \left[ \left( e^{r_{1,2}} - 1 + e^{-\beta T} \right) e^{+\beta T} \right].$$
 (7.7)

The first root of Eq. (7.5) is  $r_1 = 0$  and a rough approximation for the second root yields

$$0 < r_2 < \frac{\beta T}{e^{\alpha} l^T - 1} \tag{7.8}$$

[no difficulties are encountered in direct numerical solution of Eq. (7.7) for specified values of  $\alpha_l T$  and  $\beta T$ ].

If the initial value is r(t) > 0, we find that after a sufficiently large number of steps  $(N \rightarrow \infty)$  the function r(t + NT) tends to  $r_2$  and the solution of r(t), which describes the trailing edge of a giant pulse, can be found by expanding the difference equation (7.6) in the vicinity of  $r_2$ . On the other hand, the initial values of r(t)-corresponding to the leading edge of the giant pulse, when the (7.10)

depletion of the inversion is not significant—are close to zero, i.e., to the first root  $r_1 = 0$  and they can also be found by expansion of the initial equation.

When these points are taken into account it is found that the function r(t) in the presaturation stage is of the form

$$r(t) = e^{(\beta - \alpha_1)^t} s_0(t), \qquad (7.9)$$

where  $s_0(t) = s_0(t + T)$  is a periodic function which represents the initial stage of the emission. After inversion saturation (corresponding to the trailing edge of the giant pulse) the function r(t) becomes

 $r\left(t\right)=r_{2}-e^{-\gamma t}s\left(t\right),$ 

where

$$-\gamma = \beta - \alpha_l - \frac{1}{T} r_2 (e^{\alpha_l T} - 1)$$
 (7.11)

and the periodic function  $\mathbf{s}(t)$  represents the final stage of the emission.

Proceeding as in Chap. 5, we can establish the relationship between the functions s(t) and  $s_0(t)$ :

$$s(t) = \operatorname{const} \cdot [s_0(t)]^{-\frac{\gamma}{\beta - \alpha_l}}, \qquad (7.12)$$

which yields

$$r(t+NT) = r_2 - \operatorname{const} \cdot [r(t)]^{-\frac{\gamma}{\beta-\alpha_1}}, \quad N \to \infty.$$
 (7.13)

We shall introduce the initial and final intensity profiles:

$$I_{\text{before saturation}}(t) = i_0(t) \ e^{(\beta - \alpha_1)t} \equiv e^{(\beta - \alpha_1)t} [(\beta - \alpha_1) \ s_0(t) + \frac{ds_0}{dt}], (7.14)$$

$$I_{\text{after saturation}}(t) = i(t) e^{-\gamma t} \equiv e^{-\gamma t} \left[ -\gamma s(t) + \frac{ds}{dt} \right].$$
(7.15)

The transformation of the intensity profile is found from Eq. (7.13):

$$i(t) = \text{const} \cdot i_0(t) [r_0(t)]^{-\frac{\gamma}{\beta - \alpha} t^{-1}} e^{(\beta - \alpha t + \gamma)t}.$$
 (7.16)

The last expression allows us to estimate the factor by which the intensities at two arbitrary points of the profile in one period can change during the stage of saturation of the active material. It follows from Eq. (7.16) that the ratio  $i(t_2)/i(t_1)$  differs from the ratio  $i_0(t_2)/i_0(t_1)$ by a factor not larger than  $\exp\{(\beta - \alpha_I + \gamma)T\}$ :

$$e^{-(\beta-\alpha_{I}+\gamma)T} \leq \left(\frac{i(t_{2})}{i(t_{1})}\right) / \left(\frac{i_{0}(t_{2})}{i_{0}(t_{1})}\right) \leq e^{(\beta-\alpha_{I}+\gamma)T}.$$
(7.17)

In real laser systems the upper limit of the distortions introduced by saturation is quite low:  $\exp[(\beta - \alpha_l + \gamma)T] \leq 2-3$ . In fact, even this estimate of the distortion is strongly overestimated. This means that the saturation of the active medium has little effect on the probability of pulse selection and mode self-locking derived in Chap. 6.

We have estimated above which distortions may accumulate at the trailing edge of a giant pulse as a result of saturation of the active medium. It is obvious that these distortions will be much weaker at the peak of a giant pulse and this justifies the conclusion made at the outset of the present chapter.

#### 8. ADDITIONAL EFFECTS INFLUENCING THE FINAL STRUCTURE OF THE RADIATION

In this chapter we shall consider some effects which may influence the structure of the radiation generated in a laser with a bleachable filter.

8.1. If the velocity of propagation of light pulses along the resonator depends on the frequency (due to dispersion of the refractive index), the modes are no longer equidistant in frequency. This corresponds to dispersion spreading of the pulse along the time axis. The influence of such dispersion on the operation of a laser with a bleachable filter has been pointed out earlier.<sup>[53,54]</sup> However, only the steady-state model was considered in<sup>[53,54]</sup> and the importance of the dispersion was overestimated. We shall consider the influence of the dispersion on the parameters of laser pulses by analyzing the following simple model problem.

Let us assume that an optical field in the form of a Gaussian pulse reaches a transparent dispersive medium at the point where z = 0:

$$\mathscr{E}(z=0, t) = A_0 \exp\{-i\omega_0 t - t^2/\tau_0^2\}.$$
(8.1)

We shall find what happens to this pulse when it traverses a layer of thickness l. We shall introduce the following quantitative measure of dispersion:

$$\frac{d^2k}{d\omega^2} \equiv \frac{1}{c^2} \frac{\lambda_y^2}{2\pi} \frac{d^2n}{d\lambda_y^2} \equiv -\frac{1}{v_g} \frac{dv_g}{d\omega} .$$
(8.2)

In the second approximation of the dispersion theory (the parabolic equation approximation) the field  $\mathscr{E}$  (z = l, t) will be of the form

$$\mathscr{E}(z=l, t) = A' \exp\{-i\omega_0 t + ik_0 l - [t'^2/\tau'^2] - [ib t'^2/2] + i\varphi\}, \quad (8.3)$$

ť

$$t' = t - l/v_{g}, \quad A' = A_0 \sqrt{\tau_0/\tau'}, \quad (8.4)$$

$$\tau' = \tau_0 \, V \, 1 + 4l^2 \left( \frac{d \, n}{d \omega^2} \right)^2 \tau_0^{-4} \,, \tag{8.5}$$

$$b = 4l \frac{d^2k}{d\omega^2} (\tau')^{-2} \tau_0^{-2}, \qquad (8.6)$$

$$\varphi = \frac{1}{2} \arccos\left(\frac{\tau_0}{\tau'}\right)^{\frac{1}{2}\frac{d^2k}{d\omega^2}} \left| \frac{d^2k}{d\omega^2} \right|. \tag{8.7}$$

It follows from Eqs. (8.3)–(8.7) that the dispersion results in spreading of the Gaussian pulse and such spreading depends strongly on the initial pulse duration  $\tau_0$ , i.e., on its spectral width  $\Delta \omega_0 = 2/\tau_0$ . In our case, the dispersive medium is linear and it does not alter the spectral width of the pulse. The spreading of the pulse along the time axis is accompanied by the appearance of a phase (frequency) modulation characterized by

$$\frac{d\omega}{dt} = b = 4l \frac{d^2k}{d\omega^2} (\tau')^{-2} \tau_0^{-2}.$$
(8.8)

The influence of the dispersion on the operation of a laser with a bleachable filter can be estimated by substituting into the above formulas the initial duration of the pulse  $\tau_0$  and some effective length *l*. This length can be expressed conveniently in terms of the distance  $l_0$  traveled by the pulse in the dispersive medium in one resonator period and the effective number of passes Q:

$$l = l_0 Q. \tag{8.9}$$

We shall now estimate the values of Q and  $\tau_0$ . Since the amplification process is linear and fluctuations before the bleaching stage are of Gaussian type, it follows that the phases of different modes (spectral components) are independent. Therefore, the dispersion-induced shifts of these phases in the linear stage have no influence on the statistical properties of the initial fluctuation pattern. Thus, we can take Q as the number of passes occurring

during the bleaching stage:

$$Q \sim Q_{\rm bl} \approx \frac{\alpha_{\rm nl}}{\beta - \alpha_l - \alpha_{\rm nl}} = p - 1$$
 (8.10)

We shall assume that  $\tau_0$  is the pulse duration after the bleaching stage (Chap. 5). The influence of the dispersive medium found in this way may be overestimated. The dispersion of glass in the  $\lambda \sim 1 \mu$  range can be taken as

$$\frac{d^2k}{d\omega^2} \sim 10^{-27} \, \mathrm{sec}^2 / \mathrm{cm} \cdot$$
 (8.11)

If we assume that  $l_0 = 20$  cm and  $\tau_0 = 2 \times 10^{-12}$  sec, we find that Eq. (8.5) yields

$$\tau' = \tau_0 \sqrt{1 + (Q_{bl} \cdot 10^{-2})^2}. \tag{8.12}$$

Hence, we can see that even if the number of passes is large,  $Q_{bl} \sim 100$ , the linear dispersion increases the pulse duration by a factor of  $\sqrt{2}$ , i.e., the influence of the dispersion during the bleaching stage is slight.

This estimate is based on a very rough model: it overestimates the influence of the dispersion. In fact, at the stage when the dispersion of glass becomes important, the spectral width of a pulse  $\Delta \omega_1$  has not yet increased because of the nonlinear effect of the filter. However, the inadequacy of our model is much less important than the error resulting from the inaccuracy of the values of  $Q_{bl} = p - 1$  and  $\tau_0$  of a given laser. For this reason, we shall not go into the influence of the dispersion in greater detail.

8.2. Self-modulation of the radiation frequency should occur in the process of propagation of a pulse along a medium in the laser resonator if the field intensity in the pulse is sufficiently high (see<sup>[19,20]</sup>). This effect has been carefully studied for the case of self-mode-locked lasers<sup>[21]</sup> and the reader is referred to this paper for details. We shall just give the simplest estimates.

Self-modulation of a pulse is a result of an additional phase lead in the field of a high-power wave

$$\delta\varphi(z, t) = z \frac{\omega_0}{c} n_2 \left| E_0 \left( t - \frac{z}{v} \right) \right|^2, \qquad (8.13)$$

where  $n_2 |E|^2$  is a nonlinear correction to the refractive index (known as the reactive nonlinearity). The quantity  $n_2 |E|^2$  can be transformed conveniently to the form  $n_2 |E|^2 = h_2(n_0 c |E|^2/8\pi)$ . For glass, the order of magnitude of this correction is  $n_2 \sim 10^{-13}$  cgs esu,  $h_2 \sim 10^{-9}$  cm<sup>2</sup>/MW. Broadening of the spectrum as a result of self-modulation is of the order of

$$\delta \omega \sim \frac{\partial \omega}{\partial t} \tau_0 \sim \frac{\partial^2 \varphi}{\partial t^2} \tau_0 \sim z \frac{\omega_0}{c} n_2 |E|^2 \tau_0^{-1}.$$
 (8.14)

This broadening becomes of the order of the initial spectral width of the pulse  $\Delta \omega = 1/\tau_0$  at the point

$$z \sim z_1 = \frac{c}{m_1} [n_2 | E_0|^2]^{-1}.$$
 (8.15)

Substituting the power density  $\sim 10^3$  MW/cm<sup>2</sup>, we find from Eq. (8.15) that  $z_1 \sim 10$  cm. This means that appreciable frequency self-modulation may appear during just one pass through the laser resonator.

The dispersion and the reactive nonlinearity give rise to a time dependence of the frequency (frequency modulation) and for the majority of media (such as glass) we have  $n_2 > 0$  and  $d^2n/d\lambda_0^2 > 0$ . Therefore, these two effects should result in changes with the same sign of  $d\omega/dt$ :

In this connection, we must mention the work of Treacy,<sup>[22]</sup> who measured experimentally the value of  $d\omega/dt$ . He found that  $d\omega/dt$  was positive, which was not in conflict with the proposed mechanism of frequency modulation. Moreover, in a separate investigation Treacy<sup>[23]</sup> compressed phase-modulated pulses along the time axis by a system of diffraction gratings which simulated a medium with  $d^2n/d\lambda_0^2 < 0$ . The pulse duration obtained in this way could, in principle, have the minimum value set by the indeterminacy rule:  $\tau_{\min} \sim (\Delta \omega)^{-1}$ .

 $\frac{d\omega}{dt} >$ 

8.3. The combined effect of the dispersion and the reactive nonlinearity may lead to self-compression along the time axis (this is a time analog of the self-focusing effect) or to self-expansion of the pulses ( $\sec^{24-26}$ ). For most media (such as glass, etc.) the relationship between the signs of the reactive nonlinearity and the dispersion effect is such that self-expansion of the pulses should occur. It is worth quoting the simplest estimate<sup>[26]</sup> obtained for the distance  $z_2$  in which the self-expansion effect becomes significant:

$$k_2 \sim c\tau_0 \left\{ n_2 |E_0|^2 \left| \frac{d^2k}{d\omega^2} \right| c\omega_0 \right\}^{-1}$$
 (8.17)

For  $\tau_0 = 2 \times 10^{-12}$  sec and P ~  $10^3$  MW/cm<sup>2</sup>, we obtain  $z_2 \approx 2 \times 10^2$  cm, i.e., about 10 passes for  $l_0 = 20$  cm. This rough estimate shows that the self-expansion effect may be considerable. As far as we know, this effect has not yet been observed experimentally in its pure form.

8.4. In general, the central absorption frequency of a filter  $\omega_{\rm f}$  may not coincide with the frequency  $\omega_0$  of the laser radiation. Then, before the bleaching stage the filter not only absorbs but also produces a phase shift. After the bleaching stage the absorption and the phase shift are suppressed. This field-dependent phase shift also gives rise to a phase (frequency) modulation of laser pulses (see<sup>[27]</sup>). An explicit expression for the law of transformation of the field profile during the filter bleaching stage can be obtained—taking into account this effect—by using the general method developed in Chap. 5 (this expression is obtained in<sup>[27]</sup> by a different method).

The law of transformation of the field profile is of the form

$$(t) = \mathscr{E}_{0}(t) | \mathscr{E}_{0}(t) |^{(p-1)\left(1+i\frac{\omega_{0}-\omega_{f}}{T}\right)} \cdot \text{const;}$$
(8.18)

here, as in Chap. 5, we shall take  $p - 1 = \alpha_{nl}/(\beta - \alpha_l - \alpha_{nl})$ ;  $\Gamma$  is the rate of relaxation of the level populations in the filter. This transformation leads to the appearance of a phase modulation in the final profile. The corresponding additional broadening of the spectrum,  $\Delta \omega_f$ , is

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$$\Delta \omega_{\varphi} \sim \Delta \omega_{A} \left| \frac{\omega_{0} - \omega_{f}}{\Gamma} \right|, \qquad (8.19)$$

where  $\Delta \omega_A^{-1}$  is the duration of pulses in the field profile after the bleaching stage. In this case, the sign of  $d\omega/dt$  is the same as the sign of  $(\omega_0 - \omega_f)$ .

8.5. Bleachable materials used at present have a finite relaxation time of the bleached state. Relatively little work has been done on the relaxation process but it is known that it depends on the nature of the material.

A typical relaxation time is  $\tau_{\rm f} \sim 10^{-11}$  sec. The validity of the theory presented in Chaps. 5 and 6 in the case of a real laser depends on the ratios  $\tau_{\rm f}/\tau_1$  and  $\tau_{\rm f}/\tau'$ . Here,  $\tau_1 = (\Delta \omega_1)^{-1} = (m \Omega)^{-1}$  is the duration of the peaks in the original fluctuation pattern which is established before the onset of bleaching and  $\tau' = \tau_1/\sqrt{p}$  is the duration of the peaks which would have been generated in the bleaching of an ideal instantaneous-response filter. It is obvious that when the inequalities

$$\frac{\tau_{\mathbf{f}}}{\tau_{\mathbf{i}}} \ll \mathbf{1}, \quad \frac{\tau_{\mathbf{f}}}{\tau'} < \mathbf{1} \tag{8.20}$$

are satisfied, all the results obtained in Chaps. 5 and 6 are fully applicable. The situation described by the inequalities (8.20) represents a relatively narrow initial spectrum  $\Delta \omega_1 = \tau_1^{-1}$  and it may result from frequency-dependent resonator losses (see, for example, a report of an experimental investigation given in<sup>[28]</sup>). If

$$\frac{\tau_{f}}{\tau_{1}} \leqslant 1, \quad \frac{\tau_{f}}{\tau} > 1, \tag{8.21}$$

the duration of each peak will be reduced by a factor which is  $\sqrt{p}$  smaller than in the previous case. On the other hand, all the statistical results relating to the selection of the strongest of m initial pulses remain in force.

Finally, a situation differing strongly from the preceding one appears if

$$\frac{\tau_{f}}{\tau_{t}} \gg 1. \tag{8.22}$$

An exact theory of the effect of a bleachable filter is not yet available for the case when  $\tau_f \gg \tau_1$ . Obviously, when the inequality (8.22) is satisfied, the time dependence of the radiation will have two characteristic time scales. Some experiments<sup>[29-31]</sup> employing the twophoton method for recording the time characteristics of the laser radiation have established the existence of two characteristic time scales in the correlation of the intensity (see also<sup>[65,66]</sup>). Bradley et al.<sup>[31]</sup> have suggested that groups of peaks with a group duration  $\sim \tau_f$ are selected from the initial fluctuation pattern and that a fine structure in the form of peaks of approximately the same amplitude is observed within time intervals of the order of  $\tau_f$ .

It is not yet clear whether one such group is isolated or whether there can be several groups in one period. Moreover, it is not known whether the duration of such a group should be equal to the relaxation time of the filter or whether shorter groups may appear. The difficulty has been that even a weak response of the filter to fast changes in the intensity may give rise—because of the large number of passes of the radiation through the filter—to a considerable discrimination between the various peaks within the group and to a reduction of the duration of the group compared with  $\tau_{\rm f}$ .

8.6. In a standing-wave resonator the thickness  $l_2$  of a cuvette containing a bleachable filter material may be of importance. The influence of this thickness is, to some extent, analogous to the influence of the finite filter relaxation time  $\tau_f \sim l_1/v$  (see<sup>[31]</sup>).

If such a cuvette is placed at a considerable distance  $l_2$  from a mirror and not next to it, some special effects may be observed. The appearance of several pulses, separated by time intervals  $\Delta t = 2l_2/v$ , during one per-

iod becomes more likely. This happens because the field of one pulse causes partial bleaching of the filter and this improves the conditions for the development of a second pulse, separated by  $\Delta t = 2l_2/v$  from the first. These effects have been considered, for example, in<sup>[4]</sup>.

8.7. When the field amplitude in a pulse is large, nonlinear effects such as self-focusing, many-photon absorption, etc. may become important. The losses due to these effects increase with increasing amplitude of the pulse in question and, therefore, they may strongly impair the time structure of the radiation, giving rise to the appearance of many pulses in one period. Since these effects depend very strongly on the actual parameters of a laser, we shall not make any estimates but simply refer the reader to the original paper describing experimental investigations.<sup>[32]</sup>

8.8. In Chaps. 5-7 we have assumed that the field emerging from a laser is a quasiperiodic function of time. This means that the time structure of the radiation does not vary within a given period and that the total field amplitude, which increases at the beginning of a giant pulse and decreases at the end, varies slowly with time.

The dispersion, frequency self-modulation, and self-expansion (self-compression) effects all disturb the quasiperiodicity of the output radiation. The influence of these effects accumulates during successive passes of the pulses through the active medium in the resonator. Therefore, the phase modulation of the radiation resulting from the reactive nonlinearity  $n_2 |\mathbf{E}|^2$  and appearing at the peak of a giant pulse may vary considerably from one period to another (see<sup>[21]</sup>).

The combined effect of the dispersion and of the frequency self-modulation may result in changes not only of the phase of the field but also of the time structure of the amplitude pattern |E(t)| from one period to another. We are speaking only of changes in the fine time structure of the pulses: neither the dispersion nor the frequency self-modulation can produce changes in the intensity envelope considered on a large time scale. The nonperiodicity of the fine time structure of the radiation intensity was observed in<sup>[33]</sup>. In this investigation the two-photon method (Chap. 9) was used to show that the correlation function of the intensities  $\psi(\tau)$ 

~  $\int I(t)I(t + \tau)dt$  has a very sharp peak at  $\tau = 0$ , whereas the peak of  $\psi(\tau)$  at  $\tau = T$  is weaker and spreads over a longer time interval.

The nonperiodicity of the fine time structure of the radiation may be manifested also in various nonlinear radiation conversion processes (for example, in the conversion involving the four-photon interaction,  $see^{[34,35]}$ ).

Thus, in some cases, the time structure of the laser radiation may depend strongly on the influence of the effects considered in the present chapter.

#### 9. TWO-PHOTON METHOD FOR RECORDING THE TIME STRUCTURE OF THE RADIATION

Several difficulties are encountered in comparisons of the theories of lasers with bleachable filters with the experimental results. The main difficulty is that there is as yet no apparatus which would have sufficiently high time resolution. The width of the spectrum of a luminescence line of an active material, which is  $\sim 1-100 \text{ cm}^{-1}$ , leads us to expect (at least in principle) pulse durations of  $\sim 5 \times 10^{-12}-5 \times 10^{-14}$  sec. Direct measurements of pulses of such durations are impossible at present. Recent image converter studies<sup>[36,58,59]</sup> have made it possible to reach an exceptionally short resolution time of  $10^{-11}$  sec. When the first reports of lasers with mode self-locking appeared in the literature (1966), the time resolution of the apparatus used in direct recording of the radiation was an order of magnitude poorer.

In view of this, an indirect two-photon method for recording the time structure of the laser radiation was suggested in<sup>[38]</sup>. The method is simple and has been used extensively. However, this method allows us to determine only a characteristic time interval in which the intensity of the investigated radiation undergoes a change but it does not allow us to find whether only one radiation pulse is emitted in a period.

Unfortunately, for a long time the theoreticians and experimentalists investigating the self-locking of laser modes had been overestimating the capabilities of the two-photon method. Unjustifiable reports of full modelocking and high powers had been made on the basis of the two-photon method. In view of the fact that this method is still widely used, we shall show (following the reports given  $in^{[39-42]}$ ) what information about laser radiation can be obtained by this method.

The standard form of the two-photon method for recording the time structure of the radiation is as follows. The radiation being investigated is split into two beams of the same intensity. These beams are directed so that they reach a cuvette containing a dye solution (usually rhodamine 6G) from opposite directions. The dye is selected so as to ensure that the luminescence appears in it only when two photons of the laser radiation are absorbed simultaneously. The measured quantity is the intensity of the luminescence plotted as a function of the coordinate directed along the axis of the two beams.

The idea behind the method is that the intensity of the luminescence excited by two-photon absorption is proportional to the square of the intensity of the exciting radiation:

$$H_{\text{lum}}(x) = \text{const.} \int I^2(x, t) dt.$$
(9.1)

Therefore, at the center of the cuvette, where the difference between the arrival times of the pulses from the first and second beams is zero, the intensities of the pulses from two beams should be added and this should enhance the two-photon luminescence. At those points in the cuvette where the pulses of the photons from the two beams arrive at different times, the luminescence intensity should be lower. Thus, the presence of a luminescence peak of width  $\sim \Delta x$  at the center of the cuvette can be used to draw the conclusion that the time structure of the radiation includes peaks with a characteristic scale  $\Delta t \sim 2\Delta x/v$ , where v is the velocity of light in the cuvette.

A more rigorous analysis should make allowance for the fact that we must add not the intensities but the fields of the waves traveling in opposite directions and that a standing wave is formed where the pulses meet. Such an analysis ( $see^{[39]}$ ) yields the following result. If the two waves traveling in opposite directions have the FIG. 4. Time dependence of the intensity during a period I(t) and the trace of the two-photon luminescence brightness  $\Psi(\tau)$ . It is evident that quite different functions I(t), shown in parts a and b, give practically identical two-photon luminescence traces. If the resolution time of the oscillograph  $t_r$  is greater than the interval occupied by the group of pulses in case b, the same oscillograms are obtained in both cases.



same envelopes I(t), the luminescence at the point x should be

$$\Psi(\tau) = A \left\{ 2 \int_{0}^{t} [I(t)]^2 dt + 4 \int_{0}^{t} I(t) I(t+\tau) dt \right\}.$$
 (9.2)

The distance from the center of the cuvette x is related linearly to the difference between the times:  $\tau = 2x/v$ . Consequently, the value of  $\Psi(\tau)$  can be measured only in relative units.

It is convenient to introduce a special symbol for the time correlation of the intensity function:

$$\psi(\tau) = \frac{1}{t_{\text{tot}}} \int_{0}^{t_{\text{tot}}} I(t) I(t+\tau) dt; \qquad (9.3)$$

Eq. (9.2) then becomes

$$\Psi(\tau) = At_{\text{tot}} \{2\psi(0) + 4\psi(\tau)\}.$$
(9.4)

We have mentioned earlier that a distribution of the luminescence  $\Psi(\tau)$  with a peak at  $\tau = 0$  can be used to estimate the characteristic time scale of variation of the function I(t), which can be deduced from the width of the peak. Difficulties are encountered if the same distribution is employed to show that the incident radiation consists of one and not several pulses. Figure 4 shows two possible I(t) functions and the corresponding twophoton luminescence distributions  $\Psi(\tau)$ . We can see that only one strong peak of  $\Psi(\tau)$  is observed in case b, in spite of the fact that the incident radiation I(t) consists of several strong pulses.

It is also evident that for a given value of the total energy emitted in a time interval T the instantaneous power reached in case a is six times higher than that reached in case b.

It must be stressed that, in many cases, we cannot distinguish situations of type a and b even by means of an oscillograph. In fact, if all the short pulses in case b lie within a time interval shorter than the resolution time of the oscillograph  $t_r$  (Fig. 4b), the trace displayed on the oscillograph screen consists of one pulse in case a and in case b.

We must point out that the task of differentiating the case of one pulse from the case of several strong pulses within a time interval equal to the resonator period is not made easier by any theoretical considerations of the operation of a laser under mode self-locking conditions. On the contrary, the discussion given in the preceding chapters shows that we can have partial mode locking, corresponding to the presence of several pulses of comparable intensity in the time distribution.

The two-photon luminescence method could yield ex-

tensive information if we did not restrict it to measurements of the width of the peak in the luminescence pattern but determined the complete luminescence intensity trace. However, this trace must be determined very accurately. Unfortunately, the first quantitative measurements of such traces<sup>[46]</sup> were performed using contrast, which is a fairly rough characteristic.<sup>[60,46]</sup> It is assumed in<sup>[60,46]</sup> that there is a definite value of the luminescence intensity  $\Psi(\infty)$  which determines the brightness of the luminescence patterns at all points outside the peak ( $\tau \gtrsim \Delta t$ ). The contrast R was defined as the ratio

 $\mathbf{H} = \mathbf{f}$ 

$$R = \Psi(0)/\Psi(\infty). \tag{9.5}$$

In fact, the correlation function of Eq. (9.3) and, therefore, the brightness  $\Psi(\tau)$  are usually not constant outside the central maximum (see, for example, Fig. 4b). Therefore, the contrast R which has been discussed and measured by many workers is a quantity which represents some average trace.

We shall now give some data on the contrast obtained in several simple cases.

In the first case, the radiation represents a segment of a Gaussain random process (the radiation emitted by a laser with modes which are not locked in phase). In each specific realization (laser flash) the correlation function  $\psi(\tau)$  of Eq. (9.3) has a central maximum and a large number of weak irregularly distributed additional maxima. If the number of modes is sufficiently large, for example, m ~ 1000, we find that  $\psi(\tau)$  for some realizations will differ from this ensemble-average value by a small quantity  $\sim 1/\sqrt{m} \approx 1/30$ . Therefore, we obtain the following approximate expression

$$\psi(\tau) \approx \langle \psi(\tau) \rangle = \langle I_0 \rangle^2 \left[ 1 + |\gamma_0(\tau)|^2 \right], \tag{9.6}$$

where  $\gamma_0(\tau)$  is the normalized correlation function for the field:  $\gamma_0(0) = 1$ ,  $\gamma_0(\tau \rightarrow \infty)$  [see Eq. (6.3)]. It follows from Eq. (9.6) that the contrast of the two-photon luminescence pattern is

$$R_{1,\text{Gauss}} \approx \frac{\left[2\left(1+\left|\gamma\left(0\right)\right|^{2}\right)+4\left(1+\left|\gamma\left(0\right)\right|^{2}\right)}{2\left(1+\left|\gamma\left(0\right)\right|^{2}\right)+4\left(1+\left|\gamma\left(\infty\right)\right|^{2}\right)}\right]}=1,5.$$
(9.7)

The analogous expression for the contrast in the case when the radiation represents a Gaussian random process of the p-th order is:

$$R_{p, \text{Gauss}} = 3 \left[ 1 + \frac{2(p!)^2}{(2p)!} \right]^{-1}.$$
 (9.8)

Thus, for example,  $R_1 = 1.5$ ,  $R_2 = 2.57$ ,  $R_3 = 2.86$ ,  $R_4 = 2.96, R_5 = 2.99.$ 

Finally, we shall consider the case when the intensity of the radiation vanishes outside a certain time interval  $\Delta t$ . The time correlation  $\psi(\tau)$  of Eq. (9.3) then vanishes for  $|\tau| > \Delta t$  and the contrast of such a pulse  $R_{nl}$  becomes

$$R_{\mathbf{pl}} = \frac{2\psi(0) + 4\psi(0)}{2\psi(0) + 4\psi(\infty)} = 3.$$
(9.9)

The large difference between the values of the contrast given by Eqs. (9.7) and (9.9) has led to the impression that one can prove quite simply that only one pulse is generated in a given period. Figure 4 shows clearly that this conclusion is wrong: in both cases, the contrast of the two-photon luminescence pattern is 3, although in case b there are six pulses of approximately the same intensity in each period.

The basic inaccuracy in the definition of the contrast is the indeterminacy of the definition of  $\Psi(\infty)$ . In order to find what quantitative information on the exciting radiation can be deduced from the two-photon luminescence pattern, we must drop the contrast, which is a rough characteristic, and use more rigorous integral expressions.

In order to compare different time patterns without making a priori assumptions about their actual nature, we shall introduce (in accordance with the treatment given in<sup>[41]</sup>) the following characteristic of the radiation which we shall call the effective duration:

$$\Delta t_{\rm eff} = \left[\int_{0}^{t_{\rm tot}} I(t) dt\right]^2 / \int_{0}^{t_{\rm tot}} [I(t)]^2 dt. \qquad (9.10)$$

In many cases, the radiation in a giant laser pulse depends quasiperiodically on time, i.e., the function I(t)is of the form

$$I(t) = a(t) f(t),$$
 (9.11)

where f(t) = f(t + T) and a(t) varies little in a time interval T. It then follows from Eq. (9.10) that

$$\Delta t_{\text{eff}} = \frac{\left[\int_{0}^{t_{\text{tot}}} a(t) dt\right]^{2} \left[\int_{t_{\text{tot}}}^{t_{+T}} f(t) dt\right]^{2}}{T \int_{0}^{t_{\text{tot}}} [a(t)]^{2} dt \int_{t_{\text{tot}}}^{t_{+T}} [f(t)]^{2} dt} \equiv N \Delta t_{\text{eff}}^{(T)}.$$
(9.12)

The first factor, N, in Eq. (9.12) represents the number of periods in the envelope of the giant pulse. The second factor,  $\Delta t(\mathbf{T})$ , is the effective duration of the radiation in the time interval [t, t + T]. We shall consider only the quasiperiodic functions and use the quantity  $\Delta t(T)$  (we can show that violation of the condition of quasiperiodic-ity will have little effect on  $\Delta t_{eff}/N$ ; see<sup>[42]</sup>).

It follows from the definition

$$\Delta t_{\text{eff}}^{(T)} = \left[\int_{0}^{1} I(t) dt\right]^{2} / \int_{0}^{1} [I(t)]^{2} dt \qquad (9.13)$$

that  $0 \leq \Delta t_{eff}^{(T)} \leq T$ . We can easily see that if the function I(t) is in the form of a single rectangular pulse of duration  $\Delta t_{pl}$  during one period, we have  $\Delta t_{eff}^{(T)} = \Delta t_{pl}$ . The radiation which consists of n rectangular pulses of the same duration and same intensity in a time interval [t, t + T] is characterized by  $\Delta t_{eff}^{(T)}$  which is given by  $\Delta t_{eff}^{(T)} = n \Delta t_{pl}$  and the instantaneous value of the intensity in a pulse  $I_{inst}$  exceeds the average intensity  $\overline{I}$  by a factor of  $T/\Delta t_{eff} = (T/n)\Delta t_{pl}$ :

$$I_{\text{inst}} = \frac{1}{\Delta t_{\text{eff}}^{(T)}} \int_{0}^{T} I(t) dt. \qquad (9.14)$$

It is clear that in the case of pulses of different shape, duration, and amplitude, the quantity  ${\bigtriangleup t}_{eff}^{(T)}$  is still a measure of the instantaneous power defined by Eq. (9.14).

For a given width of the field spectrum  $\Delta \omega$  a minimum of  $\Delta t_{eff}^{(T)}$  is reached when the spectral components are completely in phase: in this case,  $\Delta t_{eff}^{(T)} \sim 1/\Delta \omega$ . The deviation of  $\Delta t_{eff}^{(T)}$  from this minimum value can be regarded as one of the characteristics of the degree of partial mode locking which is directly related to the instantaneous power. The value of  $\Delta t(T)$  can be expressed in terms of the eff

correlation function of Eq. (9.3). If we use the quasiperiodicity condition of Eq. (9.11), we obtain

$$\Delta t_{\rm eff}^{(T)} = (1/2) \int_{-T}^{T} \psi(\tau) \, d\tau / \psi(0).$$
 (9.15)

Equation (9.4), which relates  $\Psi(\tau)$  and  $\psi(\tau)$ , can also be used to express  $\Delta t(\mathbf{T})$  in terms of  $\Psi(\tau)$  measured by the eff two-photon luminescence method:

$$\Delta t_{\text{eff}}^{(T)} = \left[ (3/4) \int_{-T}^{T} \Psi(\tau) \, d\tau / \Psi(0) \right] - (T/2).$$
 (9.16)

This expression can be rewritten in the form

$$\Delta t_{\rm eff} = \frac{3}{4} \int_{-T}^{T} \left[ \frac{\Psi(\tau)}{\Psi(0)} - \frac{1}{3} \right] d\tau, \qquad (9.16a)$$

which shows that the contrast of about 3 corresponds to durations much shorter than the resonator period. The most important case is that of short effective durations such that  $\Delta t(T) \ll T$ . In this case, the right-hand side of Eq. (9.16) represents a small difference between two large quantities. It is clear that, in this case, the value of  $\Delta t(T)$  can be found only if  $\Psi(\tau)$  is measured very accurately from  $\tau = 0$  to  $\tau = T/2$ . If we assume that  $\Psi(\tau)$  is measured with an error  $\delta \Psi$ , we find that Eq. (9.16) yields

$$\delta\left(\Delta t_{\text{eff}}^{(T)}\right) = \frac{3}{2} T \frac{\delta \Psi}{\Psi(0)}.$$
(9.17)

Thus, if the two-photon method is used to find whether a single pulse of duration  $\Delta t_{pl}$  is generated in one period T, we must measure  $\Psi(\tau)$  with an accuracy  $\delta \Psi/\Psi(0) = \Delta t_{pl}/T$ . Under typical conditions,  $\Delta t_{pl}/T \lesssim 10^{-3}$ , i.e., a very high accuracy is required.

We must mention that when the two-photon luminescence pattern is recorded simultaneously with the oscillogram of the investigated radiation, the accuracy requirements may be somewhat less straight. We shall assume that an oscillogram shows that all the energy emitted in a period T is concentrated in an interval of duration  $t_r$ , which is the resolution time. Under these conditions, Eqs. (9.15) and (9.16) can be reduced to the form

$$\Delta t_{eff}^{(T)} = (1/2) \int_{-t_{T}}^{t_{T}} \psi(\tau) d\tau / \psi(0), \qquad (9.18)$$

$$\Delta t_{\text{eff}}^{(T)} = [(3/4) \int_{-t_{T}}^{T} \Psi(\tau) \, d\tau / \Psi(0)] - (t_{T}/2), \qquad (9.19)$$

and Eq. (9.17) is replaced by the following estimate of the experimental error:

$$\delta \left( \Delta t_{\text{eff}}^{(T)} \right) \approx (3/2) t_{\text{r}} \quad \delta \Psi / \Psi (0).$$
(9.20)

This means that the use of an oscillograph in the twophoton method makes it possible to reduce the requirements of the experimental precision to the level  $\delta \Psi/\Psi(0) \sim \Delta t_{\rm pl}/t_{\rm r}$ .

Under typical conditions  $\Delta t_{pl}/t_r < 10^{-2}$ , i.e., the experimental precision must still be very high.

The precision of measurements in the standard form of the two-photon method is  $\delta \Psi/\Psi(0) \sim 1$  (see<sup>[46]</sup>). Thus, this method cannot be used to determine the effective duration, i.e., it gives practically no information on the peak power of the investigated incident radiation.

These theoretical conclusions relating to the time

dependence of the radiation and the two-photon luminescence pattern were confirmed by Malyutin and Shchelev.<sup>[36]</sup> In their experiments, the laser radiation was recorded by an image converter with a resolution of  $\sim 10^{-11}$  sec and they also photographed the luminescence pattern in rhodamine 6G. The image converter measurements indicated clearly that many of the laser flashes consisted of a group of several pulses in one period and that the separation between these pulses, their number, and the relative intensities varied within wide limits. All these flashes had the same two-photon luminescence patterns.

Several recent investigations of the time characteristics of the laser radiation have been based on other non-linear effects in which some correlation function of the intensity is measured. These methods include harmonic generation,  $^{[43-45]}$  the use of optical switches,  $^{[63]}$  techniques based on the luminescence resulting from n-photon absorption (n > 2),  $^{[62]}$  and the stimulated scattering of light.  $^{[64]}$ 

These methods may have technical advantages under particular conditions but the principle of recording time characteristics is very similar to that used in the standard two-photon method. All these methods suffer from the same basic shortcoming: it is difficult to distinguish the case of a single pulse in one period from the case when many such pulses are emitted. The correlation function must be measured with a high precision  $(\sim \Delta t_{pl}/t_r)$  from  $\tau = 0$  to  $\tau = t_r/2$ . A detailed analysis of the method utilizing the luminescence resulting from the n-photon absorption is given in<sup>[61]</sup>.

Moreover, in some methods the correlation functions for different values of the arguments are measured using different laser flashes. This complicates the situation and reduces the information value of the measured quantities. In fact, a clear interpretation of such measurements can be given only if it is known a priori that different laser flashes represent radiation with fully identical time characteristics.

Summarizing our discussion, we can say that the only reliable method for investigating the time structure of the laser radiation is high-speed photography in which image converters are employed (see, for example, [5]).

#### 10. CONCLUSIONS

The basic principles of the operation of a laser with a bleachable filter are now fully understood.

A regular train of single ultrashort pulses can be produced only with some probability: the time distribution of the radiation is not always reproduced from one flash to another.

The narrowing of the initial laser radiation spectrum (when an active medium with a narrower luminescence spectrum is used or when frequency-dependent losses are introduced into the laser resonator) enhances the probability of obtaining a train of single ultrashort pulses.

The probability of obtaining a regular train of single pulses is also enhanced by operating the laser close to its threshold and by increasing the density of the bleachable filter.

Careful control of the laser operation and of the positions of various elements in the resonator should make it possible to achieve a high reproducibility of the time distribution of the radiation under experimental conditions. Direct measurements have demonstrated that one  $\leq 10^{-11}$  sec pulse per period can be generated with a reproducibility of  $\geq 95\%$ .

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