Investigation of the Shape of Radiation Pulses Emitted by a Self-Mode-Locked Laser

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An investigation was made of a ruby laser emitting a large number of axial modes. A relatively slow nonlinear absorber was used for Q switching and mode locking. The most important factor influencing the stability of operation and the shape of the pulses emitted by lasers of this type was the relationship between the reciprocal of the width of the radiation spectrum and the relaxation time T_1^a of the bleached state of the absorber. When the reciprocal of the width of the radiation spectrum was less than T_1^a, the pulses were complex and consisted of a random sequence of short spikes enclosed in an envelope whose duration was of the order of T_1^a. The results of the investigation indicated that ultrashort pulses were of fluctuation origin.

INTRODUCTION

MUCH work is currently being done on lasers operating in the self-mode-locked regime which is attained by the use of dyes bleached by powerful laser radiation. The interest in such lasers is due to the fact that the radiation they emit is concentrated in pulses of extremely short (picosecond) duration. Since the pulse duration is so short, the output power of these lasers is extremely high.

Naturally, the peak radiation power can be determined only if the pulse shape is known. Unfortunately, the resolution of modern electronic apparatus used in investigations of lasers (~ 5 psec[1]) is insufficient for a detailed study of the shape of ultrashort pulses because, in the limit governed by the width of the radiation spectrum, their duration can be as short as a fraction of a picosecond for neodymium-glass lasers. In view of this, special methods have been suggested for measuring the duration of ultrashort pulses.[2-5]

Among these methods the two-photon excitation of luminescence as a result of collision of ultrashort pulses[4,5] is especially attractive because of its simplicity. However, it has been found subsequently that this method is not sufficiently reliable because it records not only the regular ultrashort pulses emitted by a laser with a bleachable absorber but also picosecond multimode radiation pulses of fluctuation origin which are generated under Q-switching and free-oscillation conditions.[6-8] The only difference is in the contrast of the two-photon luminescence which is 3.0 for an ideal sequence of single ultrashort pulses and 1.5 for the incoherent multimode (quasithermal) radiation which consists of a random sequence of fluctuation-type intensity spikes.[9-11] Therefore, the two-photon luminescence method can be used correctly only if the contrast is measured with a very high precision.[12] However, the need for these precision measurements of the contrast destroys the simplicity and the ease of the method.

The application of an improved variant of the two-photon luminescence method[13] indicated that ultrashort pulses were of complex shape. Two variants of the pulse shape, corresponding to the experimental results, were suggested in[13]: a smooth asymmetric pulse with a short leading edge and a pulse with a Lorentzian envelope (8.1 psec at mid amplitude) consisting of fluctuation spikes (Gaussian noise of 45 cm^{-1} spectral width). The basic deficiency of the method mentioned above made it impossible to decide between these two variants. On the other hand, the current theoretical ideas on the mechanism of formation of ultrashort pulses[14-16] tend to support the second variant, i.e., that the envelope of ultrashort pulses, whose duration may amount to several picoseconds, consists of a random sequence of fluctuation spikes of duration $\tau_f \sim 1/\Delta\nu$, where $\Delta\nu$ is the width of the laser radiation spectrum.[13] According to these ideas an ultrashort pulse forms in a laser from fluctuations of the intensity of multimode radiation when this radiation passes many times through an amplifying medium and a nonlinear absorber with a short relaxation time in the bleached state. During laser emission one or several of the strongest fluctuation peaks are isolated and these are then converted to a sequence of ultrashort pulses. It is assumed that the relaxation time of the absorber T_1^a is much shorter than the durations of single fluctuations of the intensity, i.e., that the absorber response is practically instantaneous. In practice the situation is just the reverse of that assumed: for example, T_1^a for a neodymium-glass laser is at least an order of magnitude longer than the characteristic duration of fluctuation peaks, which is governed by the width of the radiation spectrum, i.e., the inequality $T_1^a \gg 1/\Delta\nu$ is usually satisfied.[13]

An allowance for the finite response time of the absorber should have the following influence on the development of ultrashort pulses. During the response (delay) time an absorber is insensitive to fast variations in the intensity and it effectively interacts with fluctuation-type radiation averaged out over the period T_1^a. This smoothing of the fluctuations reduces the chances of isolation of single ultrashort pulses and, consequently, the stability of the operation of a laser deteriorates (single ultrashort pulses in one emission period are observed less frequently). Moreover, the shape of the emitted radiation pulse becomes complex. When the absorber is bleached by a powerful fluctuation pulse it transmits weaker pulses which follow directly the strong pulse. The envelope of these weaker pulses should be of duration close to T_1^a. These theoretical considerations are supported by experimental results. A drastic deterioration of the stability of the operation of a laser emitting ultrashort pulses was found to result from a deliberately induced narowing of the emission spectrum.[17] The results reported in[18] support the idea that the radiation

of a laser emitting ultrashort pulses consists of a random sequence of pulses enclosed in an envelope governed by T_1^a.

We investigated a laser with a relatively slow absorber. The purpose of our study was to determine the influence of the inertia of the absorber response on the operation of the laser. The active medium was a ruby crystal and the nonlinear absorber was a solution of vanadium phthalocyanine in nitrobenzene. Special measurements showed that the relaxation time of this absorber was $T_1^a = 1.2 \pm 0.6$ nsec. Consequently, the resolution of an image converter camera of the FÉR-2 type[19] was quite sufficient for the study of the shape and structure of the pulses. The width of the radiation spectrum was varied by introducing selector elements into the resonator. These elements consisted of plane-parallel glass plates of various thicknesses. We investigated the following three operation regimes:

$$\text{I} - \tau_f \approx 1/\Delta\nu \ll T_1^a \ ;$$

$$\text{II} - \tau_f \approx 1/\Delta\nu \leqslant T_1^a \ ;$$

$$\text{III} - \tau_f \approx 1/\Delta\nu > T_1^a \ .$$

The results of our investigations confirmed the fluctuation origin of the formation of ultrashort pulses.

MEASUREMENTS OF THE BLEACHED-STATE LIFETIME OF VANADIUM PHTHALOCYANINE

Our nonlinear absorber was a solution of vanadium phthalocyanine in nitrobenzene. We knew that T_1^a for this solution was of the order of several nanoseconds.[20] However, we were unable to find any precise data for T_1^a and, therefore, we decided to measure this quantity experimentally.

The apparatus used in the measurement of the bleached-state lifetime of solutions of vanadium phthalocyanine in nitrobenzene is shown schematically in Fig. 1. Ultrashort ruby laser pulses were used to bleach the vanadium phthalocyanine solution enclosed in a cuvette. The self-locking of the radiation modes occurred in a cuvette filled with a solution of cryptocyanine in ethyl alcohol. The length of the resonator was such that the repetition period of the ultrashort pulses exceeded considerably T_1^a of vanadium phthalocyanine. This ensured complete relaxation of the bleached state.

Some of the radiation transmitted by a mirror R_1 reached a system of two plane-parallel mirrors R_2 and R_3 whose reflection coefficients were 85 and 95%, respectively. The separation between R_2 and R_3 was 1.5 cm. An ultrashort pulse which traversed this system degenerated into a damped sequence of spikes (Fig. 2). The degree of damping was governed by the reflection coefficients of the mirror and the repetition period was determined by the separation between the

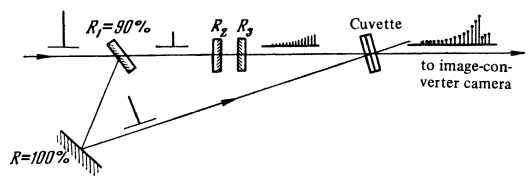

FIG. 1. Schematic diagram of the apparatus used in measurement of T_1^a of a solution of vanadium phthalocyanine.

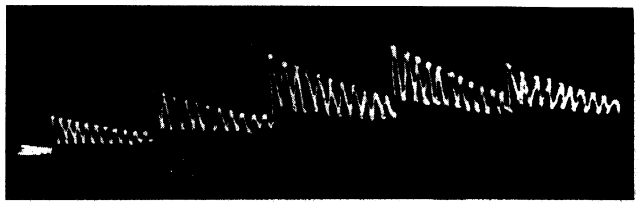

FIG. 2. Oscillogram of a damped sequence of ultrashort pulses (scanning duration 100 nsec).

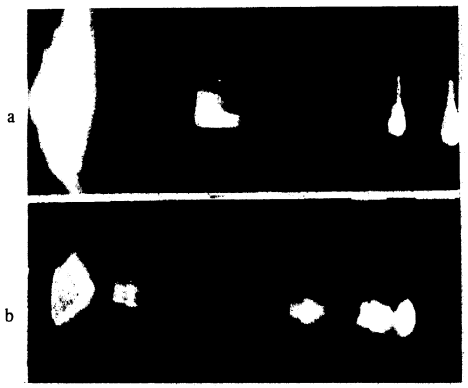

FIG. 3. Ultrashort pulses before (a) and after (b) passage through a cuvette containing a solution of vanadium phthalocyanine in nitrobenzene. The axial period was 20 nsec. The separation between pulses formed by mirrors R_2 and R_3 was 0.1 nsec.

mirrors. This sequence of spikes was then passed through a cuvette filled with a solution of vanadium phthalocyanine (the concentration of this compound was $\sim 1.3 \times 10^{16}$ cm^{-3}). The intensity of the spikes was sufficiently low not to alter the transmission in the cuvette. After some delay we injected into the same cuvette a powerful pulse which rapidly bleached the vanadium phthalocyanine could be determined from the degree of damping of the sequence of pulses which traversed the cuvette. Figure 3 shows such sequences before and after transmission through the cuvette. The sequences were recorded with an image-converter camera of the FÉR-2 type. An analysis of three records of the type shown in Fig. 3 yielded $T_1^a = 1.2 \pm 0.6$ nsec for vanadium phthalocyanine.

INVESTIGATION OF THE RADIATION EMITTED BY A LASER IN THE CASE OF MODE SELF-LOCKING ACHIEVED WITH THE AID OF A SOLUTION OF VANADIUM PHTHALOCYANINE

The apparatus employed in this investigation is shown schematically in Fig. 4. The active medium was a ruby crystal with leuco-sapphire ends cut at the Brewster angle. The active part of the ruby crystal was 240 mm long and its diameter was 12 mm. The resonator mirrors had reflection coefficients of 50 and 95%, respectively. A sufficiently large number of modes in the spectral range $1/T_1^a$ could be excited if $T_1^a \ll T = 2L/c$, where L is the length of the resonator and c is the velocity of light. Therefore, the resonator length was selected to be L = 600 cm (T = 40 nsec). Two lenses were inserted in the resonator in order to compensate for the distortions of the wave front in the ruby crystal. The resonator modes were selected with the

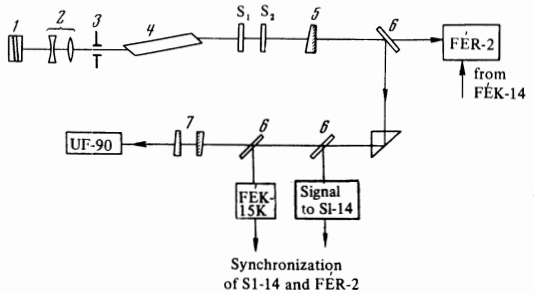

FIG. 4. Schematic diagram of the apparatus used in main experiments: 1) cuvette with 95% mirror; 2) system of lenses for compensation of wave-front distortion; 3) diaphragm; 4) ruby crystal; 5) 50% exit mirror; 6) plane-parallel plates; 7) Fabry-Perot interferometer; S_1 and S_2 are selector elements.

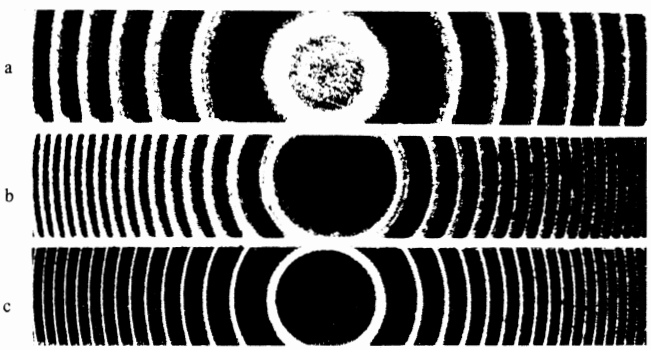

FIG. 5. Radiation spectra: a) regime I, Fabry-Perot etalon separation 12 mm; b) regime II, Fabry-Perot etalon separation 30 mm; c) regime III, Fabry-Perot etalon separation 30 mm.

help of diaphragms with apertures of 2—4 mm diameter, which were also inserted in the resonator. The output radiation spectrum could be narrowed by introducing longitudinal-mode selectors into the resonator (these selectors were plane-parallel glass plates 3 and 10 mm thick).

A part of the output radiation of the laser was directed onto the entry slit of the FÉR-2 camera in order to study the time structure of the pulses. Another part of the radiation was passed through a Fabry-Perot interferometer of the IT-28-30 type which was used to record the radiation spectrum. The Fabry-Perot interferograms were photographed with a UF-90 camera. A sequence of laser pulses could be examined on the screen of a S1-14 oscillograph with the aid of a FÉK-19 photodiode. An FÉK-14 photocell was used to trigger the FÉR-2 camera and the S1-14 oscillograph.

I. Operation Regime Corresponding to $\tau_f \approx 1/\Delta\nu \ll T_1^a$

This operation regime was achieved by taking away all the mode selectors from the resonator. The total width of the laser radiation spectrum was $\Delta\nu = 0.2—0.5$ cm^{-1}. This corresponded to $\tau_f \approx 0.1$ nsec, which was an order of magnitude shorter than the relaxation time of the absorber. The radiation spectrum obtained with a Fabry-Perot interferometer (plate separation 12 mm) is shown in Fig. 5a. The stability of the laser operation was extremely low, i.e., the appearance of a single pulse during the axial period was a rare event. An examination of the oscillograms showed that

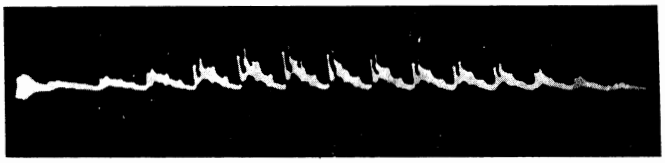

FIG. 6. Oscillogram of the radiation emitted in regime I. The pulse repetition period was 40 nsec.

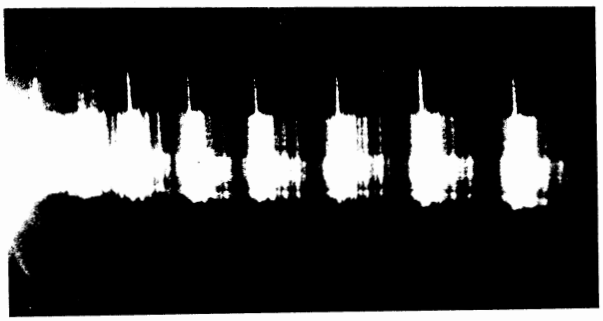

FIG. 7. Photograph of a time scan of the radiation in regime I, obtained with the FÉR-2 camera. The axial period was 40 nsec.

the output radiation was poorly modulated and the axial period was filled with sequences of short pulses. An oscillogram of this type is reproduced in Fig. 6. Streak records obtained with the FÉR-2 camera indicated that the axial period was filled with a large number of clo closely spaced pulses whose duration was of the order of 0.1 nsec. One of the best streak records is shown in Fig. 7 (a single pulse, much stronger than the other pulses, occurred once during the axial period).

A study of a large number of the streak records obtained with the FÉR-2 camera established the following features of this operation regime:

1) in any given axial period the strongest pulse was accompanied by additional pulses;

2) the duration of each pulse was governed by the total width of the radiation spectrum and the spectrum had some structure;

3) the strongest pulses and the additional random pulses formed sequences with a characteristic envelope. An analysis of a large number of photographs enabled us to establish the following features of this envelope: the leading edge was usually shorter than the trailing edge; the trailing edge was approximated satisfactorily by an exponential function with a decay constant of the order of T_1^a.

II. Operation Regime Corresponding to $\tau_f \approx 1/\Delta\nu \leq T_1^a$

This regime was achieved by narrowing the laser radiation spectrum to 0.04—0.08 cm^{-1} with the aid of selectors placed inside the resonators. The radiation spectrum recorded with a Fabry-Perot etalon (plate separation 30 mm) is reproduced in Fig. 5b. The width of the spectrum corresponded to fluctuation pulses of 0.4—0.8 nsec duration.

Figure 8 shows an oscillogram of the output radiation typical of this operation regime. The observations made with the FÉR-2 camera indicated that the probability of appearance of additional pulses in one axial period was now much lower. There were hardly any frames in which the axial period would be filled completely with

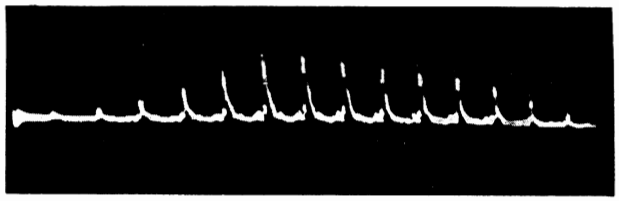

FIG. 8. Oscillogram of the radiation emitted in regime II. The pulse repetition period was 40 nsec.

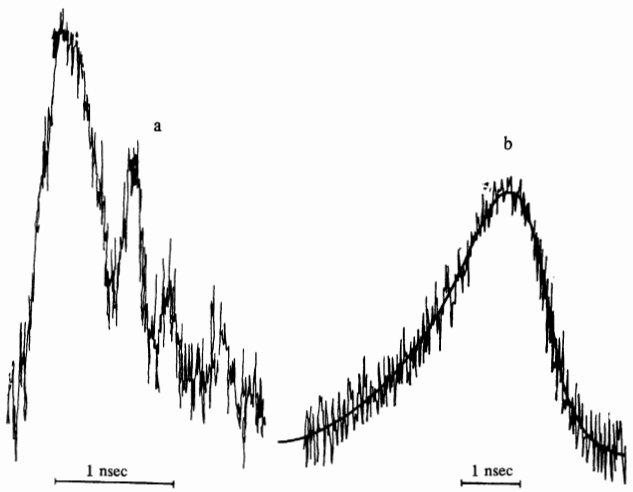

FIG. 9. Time scan of the radiation recorded with the FÉR-2 camera and subjected to a photometric analysis: a) regime II; b) regime III.

pulses. Thus, in this regime the stability of the laser operation was considerably higher. The radiation pulses were of complex shape. Figure 9a shows the results of a photometric analysis of a signal recorded in regime II with the FÉR-2 camera. The radiation was found to consist of separate pulses enclosed by an exponential envelope typical of the relaxation of the bleached state of the absorber.

III. Operation Regime Corresponding to $\tau_f \approx 1/\Delta\nu > T_1^a$

In this case the width of the radiation spectrum was reduced to less than 0.02 cm^{-1} (this was the resolution limit of our Fabry-Perot interferometer when the etalons were separated by 30 mm in air). An interferogram of such a spectrum is reproduced in Fig. 5c. The duration of the radiation pulses corresponding to the spectrum of this width should be in excess of 1.7 nsec. The stability of the laser operation was even higher than in regime II. No internal structure was found in the radiation pulses. Figure 9b shows the results of a photometric analysis of a pulse recorded with the FÉR-2 camera. The pulses now had an asymmetric shape: the duration of the leading edge was 2–4 times longer than the duration of the trailing edge. The duration of the whole pulse measured at mid-amplitude was 2.0 nsec, which was close to the relaxation time T_1^a.

Thus, a study of the operation of a laser in which modes were locked by a relatively slow nonlinear absorber enabled us to determine directly the shape of the radiation pulses. The most important factor which governed the probability of appearance of single pulses during each axial period and which determined the pulse shape was the relationship between the reciprocal of the width of the radiation spectrum and the relaxation time of the bleached state of the nonlinear absorber T_1^a. In those cases when T_1^a exceeded the reciprocal of the width of the radiation spectrum we found that random spikes appeared within the pulse envelope. The appearance of these spikes was expected because of the fluctuation nature of the formation of ultrashort pulses.

CONCLUSIONS

A laser emitting a large number of axial modes and utilizing a saturable filter with a relatively long bleached-state lifetime for Q switching and mode locking can be regarded as a model of real lasers emitting ultrashort pulses and utilizing fast absorbers. In real lasers of this type the reciprocal of the width of the radiation spectrum is usually less than the bleached-state lifetime[13] and, consequently, the shape of the pulses should be of the type observed in regime I. In other words, quasinoise pulses of duration of the order of the reciprocal of the width of the radiation spectrum should be enclosed in an envelope of duration of the order of T_1^a. Naturally, some features of the operation of real lasers cannot be described by this model. These are the features which are associated with the short durations and high powers of ultrashort pulses, such as the frequency modulation due to dispersion[21] (known as chirping) and the effects associated with changes in the refractive index in strong light fields.[22] These effects are manifested mainly in the last stage of the development of ultrashort pulses and, therefore, we may assume that our model describes correctly the earlier stages. The frequency modulation should result in some broadening of the pulses enclosed in an envelope. The results obtained in the present study suggest that ultrashort pulses produced by real lasers consist of a random sequence of spikes whose duration is longer than the reciprocal of the width of the radiation spectrum by an amount governed by dispersion and whose envelope is determined by the lifetime of the bleached state of the nonlinear absorber.

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