Direct observation of the picosecond structure of radiation pulses from a mode-locked neodymium laser

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(Submitted March 27, 1973)
Zh. Eksp. Teor. Fiz. **65**, 907–916 (September 1973)

Ultrashort radiation pulses from a mode-locked neodymium laser are investigated with a "picochron" image-convert tube, having a time resolution of 5×10^{-13} sec. A special method for recording light pulses, which excludes the effect of statistical fluctuations of the signals, is described. The total duration of the pulses is measured and the presence of a recurrent fine structure with a characteristic time of 1–2 psec is observed. The effect of various factors on the time structure of the pulses is investigated. Experiments with two types of dyes were performed with this aim in mind; the thickness of the dye layer was varied, the initial transmission remaining constant. Diaphragms of various diameters were placed in the laser cavity. The duration and shape of the pulses in various parts of the train are investigated.

INTRODUCTION

At the present time, the waveforms and the durations of light pulses emitted by neodymium-glass lasers in the mode-locking regime are being investigated. The pulse durations certainly do not exceed 10^{-10} sec, so that the standard measuring apparatus (coaxial photocells with broadband oscilloscopes, or even ordinary electron-optical recorders) do not make it possible to investigate the pulse waveform in detail.

Various methods have been proposed for indirect determination of the duration of ultrashort pulses (USP)[1-3]. These methods, based on the measurement of the correlation functions of second and higher orders, unfortunately require that definite assumptions be made concerning the temporal structure of the investigated pulse if unambiguous results are to be obtained. Were we sure that the laser in the mode-locking regime emits simple single pulses, or even pulses of complicated but of definite waveform, then the use of indirect procedures would make it possible to measure the waveform and duration of the ultrashort pulses in satisfactory fashion. There are facts, however, indicating that the USP waveform is complicated and does not repeat from flash to flash or even from pulse to pulse within a single train of USP[4-6]. To investigate the USP of a neodymium laser with mode locking, it therefore becomes very important to develop direct observation methods with temporal resolution $\lesssim10^{-12}$ sec. Such methods are based on the use of high-velocity electron-optical recorders. There are published results on an investigation of a laser operating in the mode-locking regime, using an electron-optical converter with time resolution 4 psec[7]. In the present study, the USP of a neodymium laser were investigated with the aid of the "picochron" electron-optical converter with a time resolution 5×10^{-13} sec [8].

1. EXPERIMENTAL SETUP

Description of Laser

To obtain a stable regime in which a regular sequence of ultrashort pulses is emitted, it is necessary to satisfy a number of stringent experimental requirements. The laser diagram is shown in Fig. 1. To exclude the influence of cell position on the temporal structure of the radiation, a ring-laser scheme was used with careful elimination of the selecting elements from the resonator. A system of converging and diverging lenses 1 was placed inside the resonator, and adjustment of the system compensated for the thermal distortion in the active medium and ensured the unstable-resonator regime. The use of an unstable resonator made it possible to operate with one lowest-order transverse mode at a sufficiently large beam amplitude. In an unstable resonator, the excitation threshold of this transverse mode is much lower than for modes of higher order [9]. The requirements on the accuracy of selection of the pump energy at which only one transverse mode is excited are therefore less strict. It was possible to place a diaphragm 2 of diameter 2.4 or 8 mm in the resonator. The construction of the cell 3, containing a solution of a bleachable dye in nitrobenzene, made it possible to vary the thickness of the solution layer. The minimum thickness could be 10 μ. The initial transmission of the cell was 50 or 80%. The total duration of the train was approximately 1000 nsec. The pulse repetition period was 14 nsec. The total energy in the train was 0.2 J.

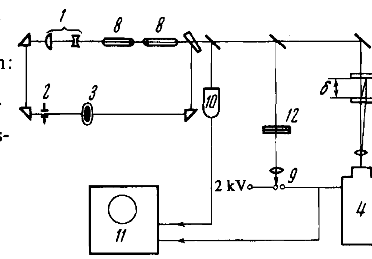

FIG. 1. Diagram of setup: 1—system of converging and diverging lenses; 2—diaphragm: 3—cell with bleachable dye; 4—picochron; 5, 6—system of two mirrors for repeated registration of the signal with distance δ between mirrors; 7—objective focusing the beam on the picochron photocathode; 8—neodymium-glass rods cut at an angle 5°, diameter 12 mm, length 300 mm; 9—discharge gap ignited by the laser radiation; 10—FÉK-15 coaxial photocell; 11—I-2-7 oscilloscope; 12—attenuating filters.

The infrared radiation of the laser was directed to the photocathode of the picochron 4 to study the temporal structure of the ultrashort pulses.

Spectral measurements of the output radiation were made with another laser of identical construction. We measured the spectrum of one pulse separated from the front part of the train with the aid of an electron-optical Pockels shutter, as well as the integral spectrum of all the pulses of the train. The spectral width of a single pulse ranged from 2 to 7 Å, and the width of the integral spectrum reached 30 Å.

Photoelectronic Recorder

It was shown in [10,11] that when fast processes are observed directly with the aid of an electron-optical converter it is possible in principle to have a temporal resolution of up to 10^{-14} sec. Of all the known electron-optical converters, the highest temporal resolution is possessed by the picochron [8]. The photoelectric recorder used in the present study, which was based on the picochron, is shown in Fig. 2. Continuous circular (elliptical) sweep of the image was effected in the input stage of the picochron with the aid of resonators 3, which were tuned to the wavelength $\lambda = 3$ cm. In addition, when the discharge gap 9 was ignited by the laser radiation, an additional single linear sweep of the image was effected with the aid of the deflecting plates 8.

The succeeding stages for the amplification of the image brightness make it possible, in principle, to photograph a minimal signal corresponding to the emission of a single electron from the input photocathode 1. If necessary, the minimal recorded signal of N_{min} primary electrons can be varied by regulating the voltages on the amplification stages.

In the experiments described below, we satisfied the condition $10 < N_{min} < 100$. At a small number of electrons N emitted from the resolved surface element of the input photocathode (0.2 mm diameter) during the resolved time, fluctuations of order $\sqrt{N}$ were observed in the registered signal, and these limited the measurement accuracy. On the other hand, at extremely large values of N_{min}, deterioration of the temporal resolution of the instrument is brought about by overloading of the input stage. Thus, high temporal resolution is obtained at the expense of a certain reduction in the accuracy of registration of the signal waveform.

The combination of continuous circular sweep and laser-triggered linear sweep in the picochron provided very important advantages. First, it was possible to trace, with a temporal resolution 0.5–1 psec, all the variations of radiation intensity in a single flash over a 30 nsec duration, i.e., a time interval exceeding the repetition period of the ultra-short pulses in the train. Second, it was possible to dispense with additional absolute calibration of the time sweep. Third, in comparison with the previously employed photoelectronic recorders, the requirements on the accuracy of the sweep triggering have been greatly simplified.

The large working time interval of the picochron made it possible to employ a special observation procedure in which the influence of the aforementioned statistical fluctuations of the signals was eliminated. To this end, one and the same primary light pulse was registered several times (up to three times), and the real structure of the pulse was taken to be only those characteristics which were traced in all the observation channels. The repeated-registration system was based on the fact that before striking the photocathode of the electron optical converter, the parallel laser-radiation beam passed through a system of two semitransparent mirrors (5 and 6 in Fig. 1). The distance between the mirrors could be varied in a rather wide range. After passing through the mirror, the initial beam broke up into several beams that were delayed relative to one another by a time $2\delta/c$. The objective 7 focused all the beams onto a single point on the input photocathode of the picochron. The repeated-registration procedure described above made it possible, first, to assess reliably whether we were dealing with the real structure of the radiation or with apparatus effects; second, it was easier to operate in the region of optimal intensity, since multiplication is accompanied by an additional weakening of the intensity; third, by varying the distance δ, it was possible to establish the direction of the image sweep.

2. EXPERIMENTAL RESULTS

Figure 3 shows photographs taken from the picochron screen in the case of multichannel registration of laser radiation. The picochron was operated with a large image-brightness gain, such that the emulsion on which the image from the output luminescence screen was photographed reached saturation. Under these conditions, the ultrashort pulse duration was measured at the base. We see that this quantity was adequately duplicated in all the registration channels and could be measured with accuracy not worse than several psec. Case a differs from case b in the value of the distance δ between the mirrors of the repeated-registration system. A comparison of the mutual phasing of the repeated signals makes it possible to determine uniquely the sweep direction indicated by the arrow (see Fig. 3c).

In the subsequent measurements, the image-brightness gain of the picochron was reduced to such a degree that the time-swept image could be measured photometrically. Figure 4 shows two examples of photometry of the image sweep in two channels. In both cases we

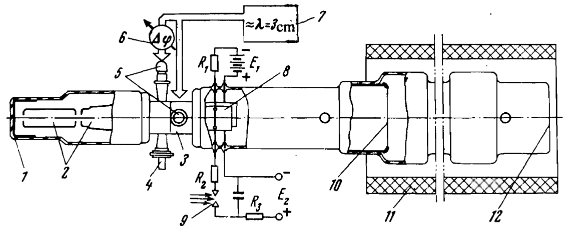

FIG. 2. Diagram of picochron photoelectronic recorder: 1–photocathode; 2–electrodes of electrostatic lens; 3–resonators of the sweep system; 4–frequency-tuning plunger; 5–microwave lead-ins; 6–phase shifter; 7–sweep generator; 8–deflecting plates; 9–discharge gap ignited by the laser; 10–luminescent screen of input stage; 11–magnetic focusing system; 12–output luminescence screen.

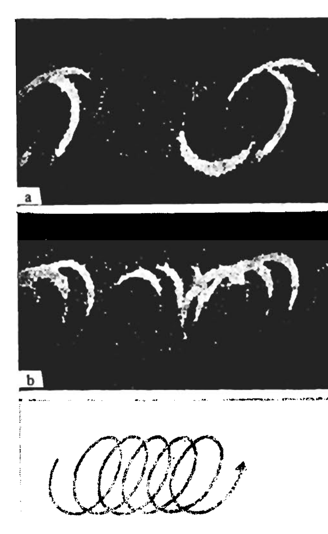

FIG. 3. Photograph of sweep of ultrashort pulses registered through three channels; a–distance between mirrors δ = 420 mm; b–δ = 410 mm; c–direction and form of the sweep.

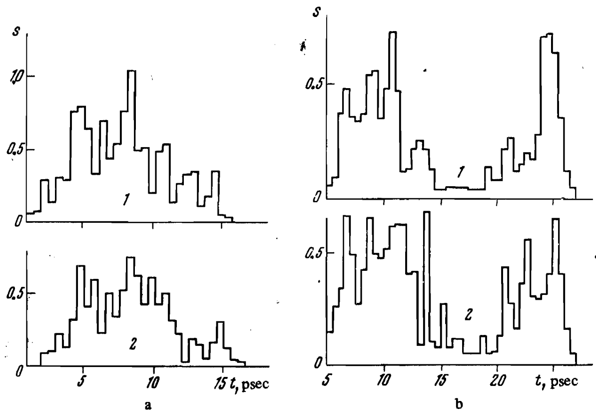

FIG. 4. Two examples of the photometry of ultrashort pulses (s—density) in registration channels 1 and 2.

see that the total duration and the general character of the signal envelope are quite well duplicated in both registration channels. The duplicated fine structure is also noted with the characteristic time ~1–2 psec. A similar picosecond structure of ultrashort pulses was observed earlier in [12].

To ascertain the influence of different factors on the form of the temporal structure of the ultrashort pulses, experiments were performed with two types of dye (No. 3955 and No. 1000); the dye-layer thickness was varied at a constant initial transmission; the initial transmission was varied; diaphragms 2.4 and 8 mm in diameter were placed inside the laser resonator; pulses in different parts of the train were investigated.

Figures 5, 6, and 7 show some of the photographs of the scanned pulses. As already noted, a distinguishing feature of the picochron operation is statistical scatter of the number of photoelectrons, so that the picochron, generally speaking, cannot determine the exact intensity profile. It is possible, however, to determine the total pulse duration at the base, to observe the presence of sufficiently clearly pronounced minima and maxima, and also to assess the presence of an internal structure in the radiation pulses.

Figures 5a, b, c, and d show examples of ultrashort pulses observed in four different laser flashes under constant external conditions. We see that even within a single measurement run the pulses differ significantly from one another from flash to flash. However, in spite of the statistical scatter of the ultrashort-pulse characteristics, it is possible to establish certain regularities for the duration and structure of the pulses as functions of the experimental conditions.

Particular attention was paid to study of the variation of the radiation-pulse waveforms along the ultrashort-pulse train. As noted above, the linear sweep of the picochron image was triggered by discharge gap 9, ignited by a laser beam (Fig. 1). Before the start and after the end of the linear sweep, the image was blocked (taken outside the field of view). The instant of sweep triggering along the ultrashort pulse train was varied by introducing different filters 12. The oscilloscope 11 displayed the ultrashort-pulse train and the pulse showing the instant of the triggering of the picochron. In this measurement series it was established that pulses at the start of the ultrashort-pulse train (200–300 nsec before the maximum) frequently have a smooth form with total duration less than 10 psec. A typical example

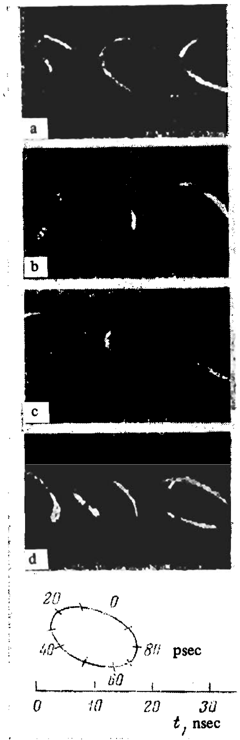

FIG. 5

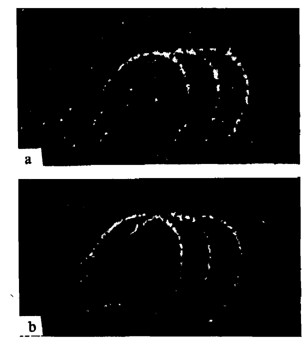

FIG. 7

FIG. 5. Pulses observed in four different laser flashes (a, b, c, d) under constant conditions: dye No. 3955, initial cell transmission 50%, cell thickness 0.01 mm, diaphragm diameter d = 4 mm. The temporal parameters of the sweep are indicated in the lower part of the figure.

FIG. 6. Ultrashort pulse sweep. Dye No. 3955, initial cell transmission 50%, cell thickness 0.01 mm, a—start of generation train, b—middle of generation train.

FIG. 7. Ultrashort pulse sweeps obtained under identical conditions but with different dyes: a—dye No. 3955, b—dye No. 1000.

FIG. 6

is shown in Fig. 6a, which shows two neighboring ultrashort pulses of one train, one of which was registered twice in succession by means of the procedure described above, while the other was registered three times. Figure 8 shows the results of photometry of all five images. Good agreement is seen between the data on the pulse waveform obtained from all five registration channels. The duration, measured at half-height, was 6 psec.

As the ultrashort-pulse train develops, the duration of the pulses increases on the average (Fig. 6b), and the waveform becomes more and more cut up, with a complicated substructure appearing. This regularity has a statistical character and is illustrated by the plots in Fig. 9, which are constructed on the basis of photometry of a large number of photographs. As seen from the diagrams of Fig. 9, the short pulses (5–10 psec) are observed only at the start of the ultrashort-pulse train, and pulses longer than 50 psec appear near the maximum of the train. Pulses shorter than 5 psec were not observed even once in our investigations.

When working with the dyes 3955 and 1000, we were unable to observe an effect of the type of dye on the form of the pulses (Fig. 7a, b). It was established that the initial dye transmission influences the temporal structure of the pulses. The change from an initial transmission 50% to 80% (the maximum transmission at which ultrashort pulses could still be produced) led to more frequent appearance of pulses shorter than 10

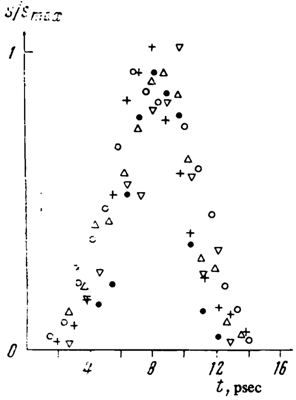

FIG. 8

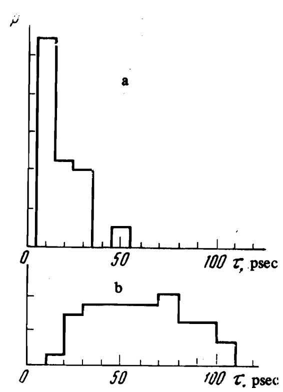

FIG. 9

FIG. 8. Results of photometry of five images of a single pulse in Fig. 6a. The ordinates represent the normalized photographic density. △, ○, +, ▽, ● pertain to the five different images of one and the same pulse.

FIG. 9. Statistical distribution of P of ultrashort pulses with respect to the duration τ at the start of the generation train (200-300 nsec prior to the train maximum (a) and near the train maximum (b)).

psec. Pulses longer than 50 psec were not observed in this case at all.

The shortest of the pulses registered by us (5 psec) was also observed at this initial transmission. In a series of experiments with a cell thickness 0.5 mm and with initial transmission 80%, we did not observe a noticeable influence of the diameter of the diaphragm 2 (Fig. 1) on the ultrashort pulses when this diameter was changed from 8 to 2 mm.

When the cell thickness was decreased to 10 μ at a fixed initial transmission, no significant shortening of the total duration of the ultrashort pulses was observed. In this case, however, pulses longer than 40 psec were very rarely observed. When the threshold gain in the laser was exceeded, and also in the case of insufficient pauses between flashes, the entire generation picture became worse. The pulses became longer, and additional satellites appeared in the axial period. The pulses with lower intensity had in this case a shorter duration.

It should be noted that in both cases the pulses in the middle of the train had a complicated asymmetrical waveform (Figs. 10a, b)—a more gently sloping leading front and a steeper trailing edge.

3. CONCLUSIONS

Oscilloscopic observations with the aid of a fast photocell (resolution 10^{-9} sec) have shown that we obtained practically every time (with probability not less than 90%) a train of single ultrashort pulses in the period. However, the picochron investigations described above have shown that the structure and duration of the ultrashort pulses vary from flash to flash; not a single pulse shorter than 5 psec was observed, and an increase of the total ultrashort-pulse duration was observed along the train, together with appearance of a complex substructure with a characteristic time up to 1–2 psec.

At the present time, the formation of ultrashort pulses in lasers with mode locking is explained on the basis of the fluctuation mechanism[13-15]. According to these notions, the lasing picture is as follows.

At the start of lasing development, a large number of

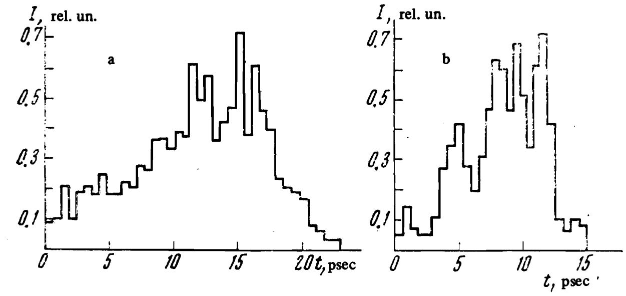

FIG. 10. Two examples of asymmetrical pulse waveforms near the maximum of the ultrashort-pulse train. The intensity I is in relative units.

axial modes is excited with a random phase distribution. Their interference produces fluctuation intensity spikes of ultrashort duration. In the nonlinear generation region, when the bleaching of the absorber takes place, the most intense fluctuation spikes become compressed and are amplified much more rapidly than the remaining less intense ones. Ultimately one or several of the most intense pulses emit the energy stored in the active region.

Within the framework of this theory, the random change in the duration of the pulses from flash to flash and the appearance of additional pulses in the axial period find a natural explanation. As to the complicated temporal structure, its appearance could also be explained, as is done in [16], in the following manner. Assuming that the dye has a finite relaxation time of the bleached state, equal to several picoseconds, one can conclude that the total duration is determined by the dye relaxation time, and that the structure consists of the fluctuation spikes. However, the experimental data of[4,5] show that the temporal structure of the ultrashort pulses is not completely reproduced from pulse to pulse in the train. Our own results also show that at the beginning of the lasing train, single pulses can appear and then be transformed into longer ones with a complicated structure (the lengthening of ultrashort pulses at the end of the train was observed with the aid of indirect methods in [6]).

It appears that single ultrashort pulses, with a duration determined by the width of the emission spectrum, are produced from the fluctuation pulses. Then, owing to the large power of the ultrashort pulse, a nonlinear interaction takes place between the light and the laser medium (the glass matrix) and leads to the appearance of nonlinear increments of the refractive index. This, in turn, causes self-focusing and self-modulation effects[17,18], which lead to a broadening of the spectrum, to frequency scanning, and, in conjunction with dispersion, to a lengthening of the pulse and to the appearance of a certain amplitude modulation. In addition, these effects are sources of additional nonlinear losses, which can alter the pulse waveforms. Indeed, weak satellites near the main pulse can become comparable with the pulse itself in intensity, and the resultant pulse becomes a complicated set of ultrashort components [19].

The influence of the nonlinear effects can be decreased by lowering the energy density in the laser resonator [19,20]. This was demonstrated in the present investigations when working with small initial absorptions of a nonlinear absorber.

In conclusion, the authors are grateful to E. K. Zavoĭskiĭ for valuable discussions, and also to V. A.

Frolov for adjusting the linear-sweep discharge gap and for help with the measurements.

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Translated by J. G. Adashko
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