

Generation of Giant Pulses from a Neodymium Laser with an Organic-Dye Saturable Filter

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There has been much recent interest in the production of high-peak power laser pulses by regeneration switching. Organic dyes have been used as saturable filters for both the ruby (cyanine dyes) and neodymium (a polymethine dye) to produce single high-power spikes.

The cyanine dye, in suitable concentration and path length, can also be used to produce a long train of slightly enhanced (gain about 10) regular Q-switched spikes from the ruby laser, but the neodymium laser can apparently only be operated in the single-pulse mode. This paper reports the discovery of saturable filter action in a common dye, rose bengal, a derivative of fluorescein, which permits the production of an extremely regular sequence of enhanced spikes from the neodymium laser. Unlike the previous dyes, the transition corresponding to the saturable absorption occurs between two excited states, not between the ground state and an excited state. The population of the lower excited state is achieved by optical pumping of the dye solution, and, by varying the intensity of the pumping flash lamp, the optical density of the filter may be altered thus changing the spacing between light spikes as well as their height.

I. INTRODUCTION

IT was first suggested by Hellwarth¹ that the laser can be made to emit fast risetime, high-intensity pulses of radiation if the cavity resonance of the laser is controlled. If the laser oscillation is inhibited until after the population inversion in the laser material has reached a maximum condition, and the regeneration of the optical cavity is suddenly switched from a low to a high value, giant pulsations of laser light which are several orders of magnitude more intense than the usual burst output may be obtained.

The methods of inhibiting the laser oscillation may be grouped into three classifications: (1) those that are mechanically active, i.e., rotating prisms or reflectors; (2) those which are mechanically passive and obtain the regeneration switching through a saturation of the absorption of a lossy medium by the laser once oscillation has begun; and (3) those which are mechanically inactive but are electrically active, i.e., Kerr and Pockels cells. Several organic dyes which may be used to Q-switch the ruby laser have been reported.²⁻⁴ These were of the cyanine type, i.e., cryptocyanine, vanadium phthalocyanine, and chloro-aluminum phthalocyanine. Recently, a polymethine dye (3,3'-diethyl-9,11; 15,17-dineopentylene-thia-pentacarbocyanine iodide) has been used to generate a single giant pulse from a neodymium laser.⁵ The purpose of this

paper is to describe a method of producing a train of giant neodymium laser pulses using rose bengal dye. The essential difference between this organic dye and those previously used is that while the Q-switching element is mechanically passive, optical pumping of the "bleachable" filter is required to populate the metastable excited state which is the initial level for the absorption.⁶ This is analogous to the uranyl glass that has been demonstrated to Q-switch the ruby laser.⁷

The purpose for using this saturable filter for the neodymium laser is connected with the use of a B-type oscilloscope presentation for a PPI laser radar system. Such a system requires a number of equally spaced spikes of uniform amplitude for target acquisition and display and the previously mentioned polymethine dye cannot be operated in the multi-Q-switch mode.

The reason that the polymethine dye cannot be operated in a multi-spike mode may be due to a large "closing" time for the dye. That is, once the filter has been bleached and the first spike of laser light has been emitted from the cavity, the total gain of the system goes to unity since the population inversion in the lasing material has been wiped out and the filter is transparent. If the flash lamp pumping the laser is still firing, the gain of the laser material will again begin to increase whereas the filter, due to various relaxation processes, may again begin to become opaque. If the filter "closes" much faster than the gain of the laser material increases, the same situation will be achieved as immediately preceded the first laser spike. That is, the filter will have a high absorption and the laser a high population inversion. If the filter "closes" slowly, however, the gain of the laser material may overcome the loss due to the

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¹ R. W. Hellwarth, in *Advances in Quantum Electronics*, edited by R. J. Singer (Columbia University Press, New York, 1961) pp. 334-341.

² P. P. Sorokin, J. J. Luzzi, J. R. Lankard, and G. D. Pettit, *IBM J. Res. Develop.* **8**, 182 (1964).

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⁶ L. A. Cross, Ph.D. dissertation, University of Michigan, 1966.

⁷ L. A. Cross and L. G. Cross, *Proc. IEEE* **54**, 1460 (1966).

saturable filter in a very short period of time so that the population inversion is not very great and the system will continue to oscillate in the normal burst fashion from then on. This type of behavior is very characteristic of the neodymium laser when Q-switched by the polymethine dye solution. (Multi-spike operation of a neodymium glass laser has been achieved by DeMaria, Stetser, and Heynau,⁸ but the total laser output envelope width was 1 μ sec or less, and the pulse-widths of the individual spikes were approximately 10^{-12} sec with a repetition period of $2-20 \times 10^{-9}$ sec. These authors interpret their results to imply as very fast "closing" time for the Eastman 9740 reversible dye solution which permits the laser to be mode-locked. On a much coarser time scale, however, it has been our experience that such a dye solution will not produce a series of large pulses spaced by several microseconds but rather that if the dye solution is sufficiently dilute, the oscillator will produce one Q-switched pulse and then continue to oscillate in its normal low-level multi-spike manner. The disparity between these two results is obvious and has not been interpreted.)

II. DESCRIPTION OF THE DYE

The dye used in these experiments was rose bengal (tetrachloro-tetraiodofluorescein) and is commonly used for biological staining. The dye used here was only 85% pure, obtained from Eastman Organic Chemicals and in most hydroxilic solvents exhibits a rather weak red fluorescence.

The parent compound for the series of compounds of which rose bengal is a member is fluoran. As substitutions are made on the six-member carbon rings of fluoran, the energy levels of the molecule are com-

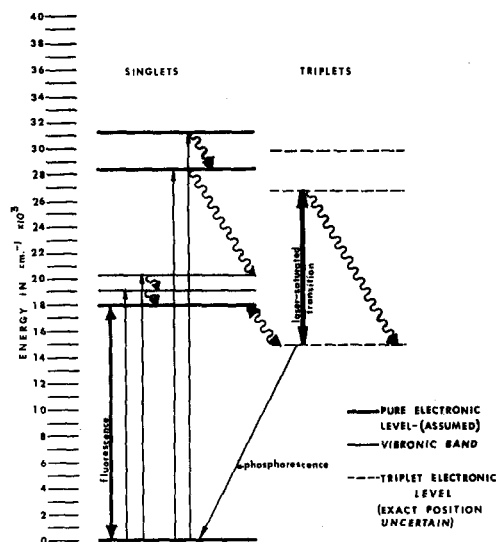


FIG. 1. Principle transitions in rose bengal.

⁸ A. J. DeMaria, D. A. Stetser, and H. Heynau, Paper 4A-7, 1966 Quantum Electronics Conference, Phoenix, Arizona.

TABLE I. Singlet and triplet band maxima for fluorescein, eosin, and erythrosin.

Substance	Fluorescence (singlet)		Afterglow (triplet)		ΔE (cm^{-1})
	λ (\AA)	ν (cm^{-1})	λ (\AA)	ν (cm^{-1})	
Fluorescein	5270	18 980	6400	15 625	3355
Eosin	5780	17 301	6500	15 384	1917
Erythrosin	5800	17 241	6700	14 925	2316

pressed bringing the first excited singlet to lower and lower energies. In order of decreasing energy-level spacing, there are: fluorescein (dihydroxyfluoran), erythrosin (tetraiodofluorescein), eosin (tetrabromofluorescein) and rose bengal (tetrachlorotetraiodofluorescein).

Figure 1 shows the energy levels of the rose bengal molecule in isopropyl alcohol solution as determined by absorption measurements. The positions of the triplet levels cannot be determined by absorption measurements since the singlet-triplet transition is highly forbidden. The singlet-triplet separation is assumed, however, to be on the order of 2000 cm^{-1} , based on the corresponding separation in fluorescein and its other derivatives. Table I lists the positions of the lowest singlet and triplet levels of fluorescein and its derivatives in solid dextrose glass as given by Pringsheim.⁹

Photo-induced absorption in fluorescein was first observed by Lewis, Lipkin, and Magel¹⁰ when a solid solution of fluorescein in boric acid was irradiated with the light from an intense mercury lamp. They observed a very broad, featureless absorption extending from 4000 to about 8000 \AA . Since the succeeding members of the fluoran series were known to have more compressed energy levels than that of fluorescein, it was decided that the last members of the series might have energy levels which were so compacted that the lower limit of the photo-induced absorption might extend down as far as 1.06μ and thus be suitable as an excited state saturable filter for the neodymium laser.

III. OBSERVATION OF THE PHOTO-INDUCED ABSORPTION IN DYE SOLUTIONS AT 1.06μ

Since it was known that photo-induced absorption, which probably corresponds to triplet-triplet transitions, could be observed in fluorescein in boric acid glass from the previously mentioned work of Lewis *et al.*, an attempt was made to observe the same effect in a glycerol solution pumped by a xenon flash lamp. The solution was contained in a 1-cm Pyrex Beckman absorption cell and suspended inside a flash head containing a linear EG&G FX-42 flash lamp, operated at about

⁹ P. Pringsheim, *Fluorescence and Phosphorescence* (Interscience Publishers, New York, 1949).

¹⁰ G. N. Lewis, D. Lipkin, and T. T. Magel, *Am. Chem. Soc. J.* **63**, 3005 (1941).

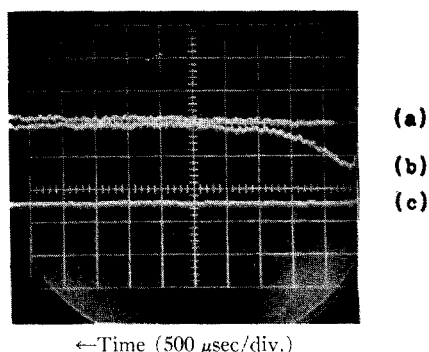


FIG. 2. Induced absorption of rose bengal dye at 1.06μ : (a) zero-absorption signal (signal light on but flashlamp not fired); (b) measured absorption signal (signal light on and flashlamp fired); (c) zero dc signal level (signal light off).

600-J input. Light from a 1600-W compact xenon arc lamp was passed through the cell, focused on the slit of a Jarrell-Ash 3.4-m spectrograph, and was detected using a Dumon 6911 photomultiplier. As is indicated by Lewis's data, the photo-induced absorption in fluorescein does begin to fall off around 8000 \AA .

Eosin dye, also dissolved in glycerol, was also tried and the expected result obtained: the photo-induced absorption does extend to longer wavelengths than in the case of fluorescein but still not as far as 1.06μ .

Figure 2 illustrates the result of the same experiment when run with rose bengal dissolved in glycerol. The upper trace represents the output of the photomultiplier when no light is passing through the sample, i.e., the xenon lamp is blocked off, and the lower trace illustrates how the signal light is diminished immediately after the flash tube is fired. (The oscilloscope sweep was initiated by the flash lamp trigger.) The sweep time on this photograph is $500 \mu\text{sec}$ per division which illustrates the extremely long lifetime of the metastable triplet state in these compounds, calculated to be about $200 \mu\text{sec}$. Fluorescein and eosin showed a similar lifetime in the spectral regions where their photo-induced absorption can be detected.

IV. Q-SWITCHING EXPERIMENTAL DETAILS

In the experiments involving the Q-switching of a neodymium laser the following laser system was used. The neodymium glass laser rod was about 20-cm long and 1 cm in diameter, one end polished flat and the other end with a roof top. To prevent the edge effects at the polished ends the end sections were enlarged (see Fig. 3). The laser rod was aligned with a resonant reflector (two clear sapphire optical flats spaced by the thickness of a flat) with a reflectivity of 65% for the laser wavelength. The laser rod was pumped with an input energy of 1800 J by two linear xenon-filled flash lamps (EG&G FX-81) in a dual-elliptical cavity. Both the flash lamps and rod were water cooled with

de-ionized water circulated through a tap-water heat exchanger. This laser system was specifically designed to produce as long a laser burst as possible and it lasted about 8 msec, beginning about 2 msec after the triggering of the flash tubes. Because of the particular type of flash tube which was available for optical pumping of the dye cell, it was not possible to design a capacitor-inductor resonant line to stretch out the duration of this flash so that it lasted as long as neodymium rod continued to oscillate. Our best effort in this direction is illustrated in Fig. 4, where it is seen that the light output from the dye-pumping flash lamp has a duration of about 4 msec with a relatively flat output for about 2 msec. The cell containing the dye solution was made of microscope slides, with parallel faces 5 cm by 4 cm spaced 1 mm apart. It was placed between the laser rod and the front resonant reflector and optically pumped by a two-turn spiral flash lamp (P.E.K. XE5-2535) with an input energy of 125 J. The laser-pumping and dye-pumping flash tubes were fired simultaneously by the same triggering unit as shown in Fig. 3. An RCA-type 922 photodiode with a sharp cut red filter was used to detect the laser radiation reflected from a white cardboard surface approximately 40 cm away from the photodiode and the same distance from the resonant reflector. The output pulses were displayed on a Hewlett Packard model 175A oscilloscope.

Three solvents for the rose bengal dye were employed: glycerol, acetone, and isopropyl alcohol (Matheson spectroquality grade). When the relative transmissions of these solvents were measured with Beckman DK-2A ratio recording spectrophotometer, the acetone exhibited the highest transmittance at 1.06μ (95% per cm) followed by isopropyl alcohol (85% per cm) and glycerol (70% per cm). (The transmission percentages of these solvents are not corrected for the reflective loss due to the glass-air and air-solvent interfaces.)

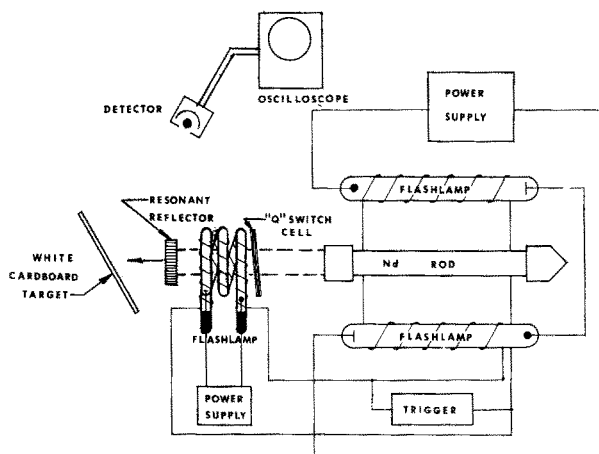


FIG. 3. Experimental arrangement of Nd laser and the Q-switch cell.

It was found that none of these liquids have any electronic energy levels in the $10\,000\text{-cm}^{-1}$ region but overtones of the vibrational structure (normally in the infrared) occur with some intensity up to about $11\,500\text{ cm}^{-1}$. Of the solvents tried which are more highly transparent at $1.06\ \mu$, none was found which was a suitable carrier for the rose bengal. The dye dissolved rather easily in acetone and isopropyl alcohol, but the very high viscosity of the glycerol necessitated the warming of the solutions to about 80°C for a half hour and vigorous stirring.

Different concentrations of dye (10^{-6} to 10^{-2} g/ml) were tested in this experiment, and the results evaluated on the basis of the ability of the filter to produce regular Q-switched spikes.

V. DISCUSSION OF RESULTS AND SUMMARY

Operation of the neodymium laser in the multi-spike Q-switched mode was accomplished for all three of the solvents mentioned before and in concentrations varying from 10^{-2} to 10^{-4} g/ml. Because an excited-state transition is involved and even at 10^{-4} g/ml the solution is opaque to the flash tube pumping radiation, the concentration is not critical. However, for all three solvents the best performance was obtained at a concentration of 10^{-3} g/ml. Acetone, because of its much higher transmission coefficient at $1.06\ \mu$, would be the most logical choice for the solvent but, because of the relatively low Q of the cavity as a whole, not much difference in performance is seen between it and isopropyl alcohol. Glycerol, because of the difficulty in preparing solutions and the inhomogeneities introduced when a sample cell was filled from a storage bottle, was soon abandoned when it was discovered that the other solvents would also work. Glycerol is also much more sensitive to contamination by atmospheric water which introduces concentration inhomogeneities in the solutions. One peculiarity of the acetone solution was noticed: The solvent vaporizes or perhaps dissolved gases are liberated and a small number of bubbles are released which rise out of the sample cell immediately after the laser is fired. This effect was not noticed for the other two solvents and does not seem to be detrimental to the laser pulse, probably because the bubbles do not reach any appreciable size during the duration of the pulse. This effect is definitely due to the absorption of laser energy by the saturable filter, since it requires the combination of both optical pumping by the flash lamp and by the laser to produce the bubbles. The results described in the rest of this section pertain to the acetone solution but the isopropyl alcohol solution behaves in essentially the same manner.

Figure 4 illustrates the significant features of the multi-spike Q-switched laser action at $1.06\ \mu$. The upper trace is the normal burst output of the laser, the middle trace is the output of the flash lamp pumping the cell containing the dye, and the lower trace shows

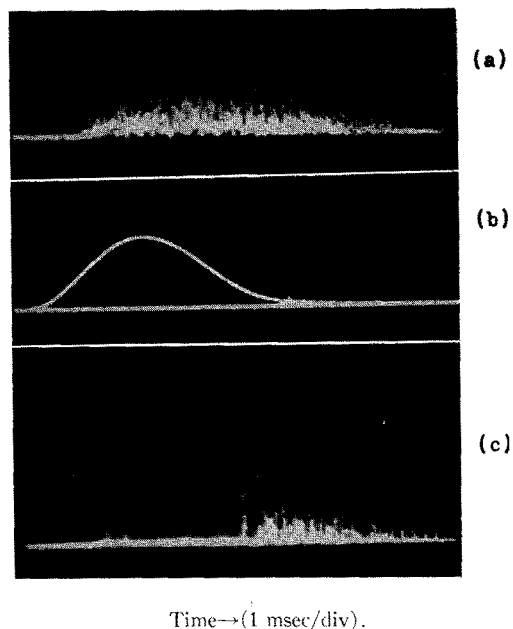


FIG. 4. Giant pulses obtained utilizing optically pumped rose bengal dye: (a) the ordinary burst Nd laser output when Q-switching cell unpumped; (b) pulse shape of the spiral flash-lamp used for pumping the Q-switch cell; (c) giant pulses obtained when Q-switch cell is pumped.

the enhancement of the peak power of the laser and the regularity of the emission spikes caused by the pumping of the dye cell. (The dye cell is also present inside the cavity during the upper trace but the cell-pumping flash lamp was not fired.) The peak power of the laser is enhanced by about a factor of 5 and the random pattern of output spikes has been replaced by a regular series whose spacing is approximately $125\ \mu\text{sec}$. The laser burst, because of the difference in the time constants of the two types of flash lamps, lasts longer than the photo-induced absorption in the saturable filter and the laser returns to its irregular spiking once the dye-pumping flash lamp has ceased to fire. It is of interest to observe the transition region between these two modes of operation and to notice that as the dye-pumping flash lamp intensity begins to diminish, the giant pulses of the laser begin to crowd together and to diminish in height. This is the type of behavior to be expected for a system of this kind.

The foregoing mode of operation was obtained by operating the laser system such that the gain of the neodymium rod was sufficient to permit it to oscillate due to the regeneration provided by reflection from its own front face. When the rod begins to oscillate, the absorption of the saturable filter is eliminated and the cavity switches from the low Q provided by the reflection from the front of the rod to the high Q provided by the reflection from the resonant reflector. Careful design of the inductor-capacitor resonant line

feeding the dye-pumping flash lamp is required to insure that the train of pulses is uniform. The small variations in the intensity of the flash lamp shown in Fig. 4 are reflected in the variation of the spacing between adjacent pulses. All efforts to use this saturable filter to generate a single spike output have been unsuccessful. Without much more sophisticated measurements than have been reported here, it is impossible to estimate the number of dye molecules which are actually participating in the photo-induced absorption and thus the amount of energy which must be supplied by the laser beam to the filter to saturate it. It is expected that electronic transitions which are not spin-forbidden would have large absorption cross sections in organic molecules of this type, but this may not be the case in this instance. It is very possible that other derivatives of

fluorescein which are closely allied to rose bengal may perform in a more satisfactory manner when only a few Q-switched spikes are desired. The construction of a system in which all undesired reflections are eliminated by the use of Brewster's angle interfaces such as that utilized in the ruby laser Q-switched by the uranyl glass⁷ would simplify the resonator and may improve the performance.

The excited-state saturable filters, of which this is the second published example, offer one major advantage over the ground state variety: The optical density of the filter may be varied at will by simply changing the pump energy to the filter-pumping flash lamp. For ground-state saturable filters this can, of course, only be accomplished by mechanically changing the filter cell length or dye solution concentration.

Formulation of Dynamic Switching in Distributed Ferromagnetic-Film Systems

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The magnetodynamic equation in the Landau-Lifshitz form has been formulated for distributed ferromagnetic-film systems, such as films with laminar magnetization or domain walls. Initial and boundary conditions are examined. The formulation lends itself easily to the numerical solution of both quasistatic- and dynamic-magnetization reversal problems, as illustrated by several examples. A number of applications for the formulation are also indicated.

I. INTRODUCTION

A DISTRIBUTED ferromagnetic-film system refers to a structure in which the magnetization orientation varies with the spatial coordinates. Some familiar examples are the various types of domain walls,¹ dual uniaxial films,² and films with laminar magnetization.³ A distinctive feature of the distributed systems is that the exchange energy is no longer zero; whereas in single domains, the exchange energy is zero.

Dynamic switching implies that the speed of magnetization reversal is of concern. Hence, the factors influencing speed such as intrinsic and eddy-current dampings must be taken into account. In the limiting case when the magnetic-field variation is so slow as to

allow magnetization variation to follow closely, the quasistatic magnetization reversal will result.

Although magnetization reversal in thin films has been extensively studied, theories developed for distributed systems have been limited to quasistatic considerations,^{1,2,4} or viscous-damping situations.³ Furthermore, very restrictive approximations, such as lumped-constant method,² small-angle theory,⁵ and Ritz method,¹ are often made to simplify the analysis. As for the analysis of dynamic switching, only single-domain configurations⁶⁻⁸ have been considered.

In this paper, the general magnetodynamic equation for distributed ferromagnetic-film systems is derived (Sec. II). In particular, the equation is applied to the films which permit variation of magnetization orientation along the direction perpendicular to the film plane. The boundary condition for such a configuration is also derived (Sec. III).

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† This paper will form part of the Ph.D. dissertation of Y. S. Lin.

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