

# A SIMPLE AND RELIABLE FLASHLAMP EXCITED ORGANIC DYE LASER

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## ***ABSTRACT***

The flashlamp-excited dye laser is recently the only type of laser capable of tunable emission throughout most of the visible spectrum. A laser using a solution of Rhodamine 6G in alcohol excited by a coaxial flashlamp is reported. The electrical energy supplied to the lamp is 45 joules. The importance of rapid pumping is considered. Constructional aspects of a suitable flashlamp assembly are described, and lasing characteristic is summarized.

## **I. INTRODUCTION**

The first unambiguously successful discovery of laser emission from organic molecules was made in 1966 by Peter Sorokin and John R. Lankard at the Thomas J. Watson Research Center of the International Business Machines Corporation. These authors used a giant pulse ruby laser to excite solutions of the dyes chloro-aluminum phthalocyanine<sup>1</sup>. Based upon the laser-pumped laser results, Sorokin, Lankard, Hammond, and Moruzzi<sup>2</sup> suggested that it is possible to obtain a dye laser by using the flashlamp excitation and estimate the flashlamp requirements. A suitable flashlamp was constructed and laser emission from solutions of several dyes was reported<sup>3,4</sup>. Hence, flashlamp excited dye lasers have since been studied in many laboratories.

During the past few years, the intense interest in dye laser research has been motivated by the dye laser's ability to provide visible coherent radiation. Although there have been numerous different schemes for obtaining the laser light, whether they were of the solid, liquid or gaseous, one of the most attractive properties of the laser is tunability. Moreover, the dye lasers are the simplest and most convenient in the visible regions of the spectrum. Dyes have been found which emit laser radiation in the spectral region from  $3450 \text{ \AA}$  to  $12000 \text{ \AA}$ <sup>5</sup>. Research in this area is currently very active.

In the present work here, we want to describe a very simple and reliable dye laser setup which is able to produce laser emission and is called flashlamp excited organic dye laser, and the basic principles of the dye laser.

We must pay attention to the attainment of very short optical pumped pulses in order to achieve flashlamp excited organic dye lasers. Both the reasons for this requirement as well as approximate determination of the actual limit on the risetime were given in section 8 of reference 2.

For the past several months we have been observing various features of the flashlamp excited dye laser. The relationship between the concentration of dye and wavelength of laser emission was studied. Rhodamine 6G was used as a dye solution in our experiment. The most intensive narrow band laser emission in the spectral range between  $5600 \text{ \AA}$  and  $6000 \text{ \AA}$  was achieved with solution around  $10^{-4} \text{ M}$  of the dye in alcohol (see appendix A).

## II. THEORY

A dye is any substance able to impart its color to other materials. However, for our purpose—organic dye laser—any organic substance absorbing in the near ultraviolet, visible, or near infrared regions of the spectrum is called organic dye. Most common dyes are solids and are dissolved in suitable solvents. In our work we use Rhodamine 6G dissolved in alcohol as the dye solution.

### A. Properties of Organic Dye Solutions.

To explain the optical absorption and emission properties of organic dyes and to introduce the discussion of the dye laser process, we use the energy level diagram shown in Fig. 1.

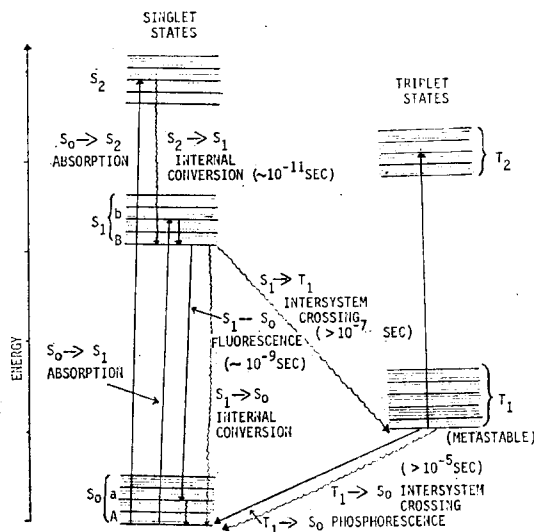


Fig. 1. Energy Level Diagram of A Dye Molecule.

There are many bound vibrational states in each electronic state and many bound rotational states in each vibrational state. The electronic ground state of the dye molecule is a singlet state, S<sub>0</sub>, which includes a range of energies determined by the quantized vibrational and rotational states of the molecule. The energy difference between neighboring vibrational states, indicated in Fig. 1 by heavy horizontal lines, is approximately of the order 1400 to 1700 cm<sup>-1</sup>. The energy difference between the vibrational levels is greater by a factor of approximately 100 than that between the rotational levels, designated by the lighter lines. Therefore, the rotational levels provide a near continuum of states between the vibrational levels.

Each electronic state of the dye molecule consists of a similar broad continuum. Optical transitions between these continua then give rise to characteris-

tic broad absorption and emission spectra. The vibrational transitions determine the wavelength region of the spectrum and the rotational transitions determine the separation of the lines. The first and second excited singlet states are designated as  $S_1$  and  $S_2$  and excited triplet states as  $T_1$  and  $T_2$  in Fig 1.

As the first step of the laser process the dye molecules are excited from the lowest level of the ground singlet  $S_0$  state to higher vibrational-rotational levels of  $S_1$  state by absorbed energy from light. This process is indicated by the transition from state A to state b ( $A \rightarrow b$ ). Most of the excited molecules decay nonradiatively to the lowest vibrational level B of the  $S_1$  state ( $b \rightarrow B$ ). It is also possible to excite dye molecules from the lowest level of the ground state  $S_0$  to the second excited singlet  $S_2$  state by absorption of energy. However, the nonradiative and very rapid decay from  $S_2$  to  $S_1$  state occurs for most organic dye solutions in about  $10^{-10}$  to  $10^{-11}$  second. This spontaneous emission (fluorescence) is called internal conversion.

The dye molecule in  $S_1$  state can return to  $S_0$  state by emitting a photon of light ( $B \rightarrow a$ ) whose energy is less than of the absorbed light; this process is called fluorescence. Because a is a higher vibrational-rotational state of  $S_0$ , hence, the process is terminated by the radiationless decay  $a \rightarrow A$ . The energy difference between absorption and emission processes is taken up by the nonradiative process  $b \rightarrow B$  and  $a \rightarrow A$ . Therefore, this spontaneous radiative process (fluorescence) is shifted to longer wavelengths from the absorption.

The transitions between singlet states ( $S_2 \rightarrow S_1$ ,  $S_1 \rightarrow S_0$ ) are spin-allowed processes; the fluorescence ( $S_1 \rightarrow S_0$ ) is governed by the lifetime  $\tau$  of state B. For organic dye molecules  $\tau$  is typically approximately  $5 \times 10^{-9}$  second. In contrast, the corresponding decay time for solid-state and inorganic laser materials is orders of magnitude slower—about  $10^{-7}$  to  $10^{-3}$  second.

Dye molecules in the singlet  $S_1$  state may undergo a nonradiative process to a lower lying triplet state  $T_1$ . This process, known as intersystem crossing, proceeds at the intersystem crossing rate  $K_{ST}$ . The lifetime of the triplet state,

$\tau_T$ , is generally much longer than fluorescence lifetime  $\tau$  since the triplet-singlet transition is spin-forbidden. The decay process  $T_1 \rightarrow S_0$  may be radiative or nonradiative. The radiation accompanying the process is called phosphorescence.

The laser emission also takes place from the stimulated transition  $B \rightarrow a$ , provided that conditions necessary for optical gain are met. Without gain, only spontaneous emission (fluorescence) occurs. From the above discussion, we know that fluorescence, internal conversion, and intersystem crossing are spontaneous transitions. When dye molecules in  $S_1$  state return to  $S_0$  state, these three types of spontaneous transitions compete with the radiative process that produces the laser light, and reduce the fluorescence quantum efficiency  $\Phi$ , that is defined as the ratio of the number of fluorescent photons emitted to the number of excitation photons absorbed by a large number of dye molecules. It is clear that a high fluorescence quantum efficiency is desirable in a dye system. For most laser dyes  $\Phi$  lies between 0.5 and 1.0. The actual value of  $\Phi$  for a given dye depends upon the solvent, the temperature, and other experiment conditions.

## B. Optical Gain in Dye Solution.

As previously stated, dye molecule in the excited singlet  $S_1$  state may relax via a radiative process to  $S_0$  state; this stimulated transition  $B \rightarrow a$  produces laser emission. The gain constant  $\alpha$ , which is defined by the gain equation,

$$\alpha = -\frac{1}{L} \ln \left( \frac{I}{I_0} \right).$$

$I/I_0$  is the ratio of the light intensity transmitted by a dye solution of path length  $L$  cm to the intensity of an incident probe light beam. The gain constant has dimension of molecules per cm and is the absorption cross section times the dye molecules populations.

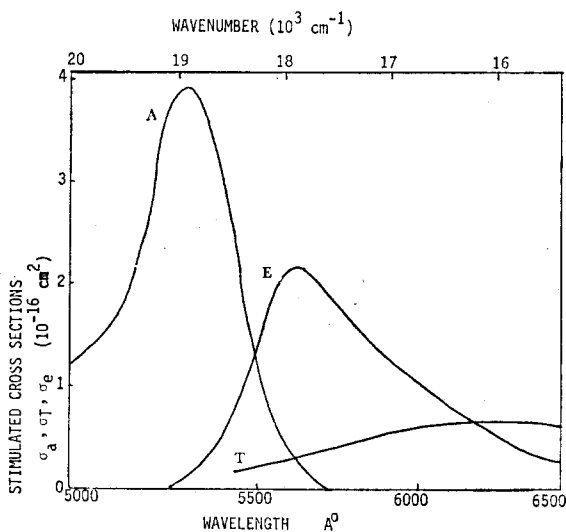
Let us consider only  $S_0$  and  $S_1$  states for the moment; the gain equation can be expressed as

$$I(Z) = I_0 e^{\alpha Z}, \quad (2)$$

and gain constant  $\alpha$ , gain per unit length at wavelength  $\lambda$ , is given by

$$\alpha(\lambda) = N_1 \sigma_e(\lambda) - N_0 \sigma_a(\lambda). \quad (3)$$

Here,  $N_1$  and  $N_0$  are the populations, number of molecules per unit volume, of the  $S_1$  and  $S_0$  states, and  $\sigma_e(\lambda)$  and  $\sigma_a(\lambda)$  are the emission and absorption cross section, at wavelength  $\lambda$ , originating from those states respectively. Owing to Boltzman distribution of populations within the continuum of each electronic state,  $\sigma_e(\lambda)$  is not equal to  $\sigma_a(\lambda)$  in organic solutions. In Fig. 2, we find the cross section as a function of wavelength for the laser dye Rhodamine 6G. As the wavelength of emission increases,  $\sigma_e(\lambda)$  can be greater than  $\sigma_a(\lambda)$ , it implies that the amplification can be easily achieved with only a small fraction of the molecules in the  $S_1$  state. In other words, the long wavelength of the dye emission makes it possible to obtain amplification even when  $N_1 \ll N_0$ .



**Fig. 2. Singlet Absorption, Emission, and Triplet Absorption Cross Sections for Rhodamine 6G.**

The absorption cross section is a readily measured quantity, and the emission cross section is obtained from the spontaneous emission (fluorescence) spectrum by relationship<sup>6, 7</sup>

$$\sigma_e(\lambda) = \frac{\lambda^4 E(\lambda)}{8\pi\tau c n^2}, \quad (4)$$

where  $n$  is the index of refraction,  $E(\lambda)$  is the true emission,  $\tau$  is the lifetime of fluorescence, for normalization,

$$\int_0^\infty E(\lambda) d\lambda = \Phi, \quad (5)$$

where  $\Phi$  is the quantum efficiency of fluorescence.

In the above description of a dye laser we neglected the other detrimental effects that may quench or even prevent laser emission. If we consider the intersystem crossing, some of the molecules in the  $S_1$  state transit into the  $T_1$  state. This process reduces the quantum efficiency. The  $T_1 \rightarrow T_2$  transition may occur due to the relatively large triplet-triplet absorption cross section. This transition usually overlaps the singlet emission and presents a loss. Similarly,  $S_1 \rightarrow S_2$  transition presents a loss too. Taking these losses into account, the gain constant at wavelength  $\lambda$  is

$$\alpha(\lambda) = N_1 \sigma_e(\lambda) - N_0 \sigma_a(\lambda) - N_T \sigma_T(\lambda) - N_1 \sigma_s^*(\lambda), \quad (6)$$

where  $N_T$  is the population of the triplet state  $T_1$ ,  $\sigma_T(\lambda)$  is the triplet absorption cross section ( $T_1 \rightarrow T_2$ ), and  $\sigma_s^*$  is the excited singlet absorption cross section ( $S_1 \rightarrow S_2$ ). In Fig. 2 the triplet cross section  $\sigma_T$  is also given for Rhodmine 6G, but  $\sigma_s^*$  is not known.

Now we consider the steady-state condition for the following derivation. We assume a dye cell with length  $L$  and cross section area  $A$ . The dye is being excited by radiation impinging on the area  $A$  and absorbed along the length, which is the lasing axis as well. In this longitudinal pumping geometry the pumping source is usually another laser. Neglecting all losses for the moment, the gain constant is

$$\alpha(x) = N_1(x) \sigma_e, \quad (7)$$

where  $x$  is measured along the laser and pumping axis. It is necessary to reach laser threshold before a lasing operation; for operation close to threshold, the rate equation for the population  $N_1$  is

$$\frac{dN_1}{dt} = N_0\sigma_{a,p}n_p(x) - N_1(x)/\tau, \quad (8)$$

where the subscript p refers to the pump wavelength, and  $n_p(x)$  is the pump photon intensity at x. If excitation takes place to the  $S_2$  state instead of the  $S_1$  state, equation (8) is still valid, provided that all the dye molecules in  $S_2$  state return quickly to the  $S_1$  state. In that case  $\sigma_{a,p}$  is the absorption cross section  $\sigma(S_1 \rightarrow S_2)$ . Of course, we suppose the relaxation process from  $S_2$  to  $S_1$  is perfect; otherwise, we need some correction factor in the first term of equation (8). After steady state has been reached, there is no rate of change in  $N_1$ , i.e.

$dN_1/dt = 0$ , so

$$N_1 = \tau N_0 \sigma_{a,p} n_p(x), \quad (9)$$

and

$$\alpha(x) = \sigma_e N_1(x) = \tau N_0 \sigma_{a,p} n_p(x) \sigma_e. \quad (10)$$

From the equation (1) and (2), we know that the total gain G from the dye cell with length L is the integration of gain constant  $\alpha$  over the length L, that is

$$G = \int_0^L \alpha(x) dx = \tau N_0 \sigma_{a,p} \sigma_e \int_0^L n_p(x) dx. \quad (11)$$

Here, we have assumed that  $N_1 \ll N_0$  and  $N_0$  is independent of x. The pump photons are being absorbed according to

$$n_p(x) = n_{p,0} e^{-\sigma_{a,p} N_0 x}, \quad (12)$$

where  $n_{p,0}$  is the input photon intensity. Substituting equation (12) into equation (11), we get

$$G = \tau \sigma_e n_{p,0} (1 - e^{-\sigma_{a,p} N_0 L}) = a n_{p,0} \sigma_e \tau. \quad (13)$$

The term in the parenthesis,  $a = (1 - e^{-\sigma_{a,p} N_0 L})$ , is the fraction of the pump power absorbed by the dye. Taking the typical values for Rhodamine 6G,  $\sigma_e \simeq 10^{-6} \text{ cm}^2$ ,  $\tau \simeq 5 \times 10^{-9} \text{ sec}$ , to achieve a total gain of 10%, and assuming that the fraction of the pump power absorbed by the dye approaches unity, we need



$$n_{p0} \simeq \frac{0.1}{10^{-16} \times 5 \times 10^{-9}} = 2 \times 10^{23} \text{ photons/sec-cm}^2.$$

From the previous discussion, we know that if the optical gain is too small, there has been no laser emission. However, from the equation (13), we find that the product of the lifetime and pumping intensity should be larger than threshold for the laser action. It means that the shorter the fluorescence lifetime, the higher the pumping intensity required. For a given peak value of  $\sigma_e$ , a broad fluorescence band results in a short lifetime  $\tau$ . Thus, the very broad bandwidth of dye laser also needs high pump intensity.

Now let us take account of the singlet and triplet losses in a more complete expression for the gain. If  $N$  is the density of total dye molecules, and  $N_0$  is the density of ground state molecules,  $N_0$  can be approximated by

$$N_0 \simeq N - an_{p0}\tau/L. \quad (14)$$

For the lowest triplet state, the rate equation can be expressed

$$\frac{dN_T}{dt} = N_1 K_{sT} - N_T/\tau_T, \quad (15)$$

where  $K_{sT}$  is the intersystem crossing rate and  $\tau_T$  is the triplet lifetime. For steady state ( $t \gg \tau_T$ ), the rate of change of triplet state is zero, i.e.

$$N_T(x) = N_1(x) K_{sT} \tau_T, \quad (16)$$

combining equation (6), (9), and (16), we obtain

$$\alpha(x) = N_0 \tau \sigma_{ap} (\sigma_e - K_{sT} \tau_T \sigma_T - \sigma_a^*) n_p(x) - N_0 \sigma_a, \quad (17)$$

then, substituting the equation (12), (14), and (17) into the definition of total gain, we get

$$\begin{aligned} G &= \int_0^L \alpha(x) dx \\ &= \tau (\sigma_e - K_{sT} \tau_T \sigma_T - \sigma_a^*) \int_0^L n_{p0} e^{-\sigma_{ap} N_0 x} \sigma_{ap} N_0 dx \\ &\quad - \sigma_a \int_0^L (N - an_{p0}\tau/L) dx \end{aligned}$$

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$$= a\tau n_{p,0} \{ \sigma_e (1 - K_{ST} \tau_T \sigma_T / \sigma_e) - \sigma_s^* \} - (NL - a n_{p,0} \tau) \sigma_a. \quad (18)$$

If we assume a constant value of cross sectional area  $A$  and a uniform power density over this area, the pump wavelength  $\lambda_p$  and input pump power in watts  $P_p$ , each input photon contains energy  $h\nu = \frac{hc}{\lambda_p}$ , and the number of in-

put photon is  $N_p = \frac{P_p}{h\nu} = \frac{P_p \lambda_p}{hc}$ ; hence, the input pump photon intensity  $n_{p,0} =$

$\frac{N_p}{A} = \frac{P_p \lambda_p}{A hc}$ , so we can express equation (18) as

$$G = (a\tau P_p \lambda_p / A hc) \{ \sigma_e (1 - K_{ST} \tau_T \sigma_T / \sigma_e) - \sigma_s^* \} - (NL - a P_p \lambda_p \tau / A hc) \sigma_a. \quad (19)$$

However, equation (19) contains most of the important qualitative features, and the quantitative differences are small.

Since the  $\tau_T$  can be large compared with the intersystem crossing decay time  $K_{ST}^{-1}$ , the quantity  $K_{ST} \tau_T \sigma_T / \sigma_e$  can be of the order of unity, even for dyes with high quantum yield ( $K_{ST}^{-1} \ll \tau$ ). For a given dye the gain value of  $K_{ST} \tau_T$  may not be achieved on a steady state regardless of the pumping intensity of the exciting radiation; that is why some dyes did not lase (see table I).

According to equation (19), the more input power we have, the more higher total gain we get. In other words, singlet losses can be overcome somewhat by sufficiently strong excitation. However, the required pumping intensities to stimulate most of the dye molecules into the  $S_1$  state are very high. Generally singlet losses limit the gain on the short wavelength side of the emission band. For instance, in all practical experimental situations, Rhodamine 6G does not lase at the peak of its fluorescence.

**Table I. lasing and nonlasing dyes**

| Dye                               | Solvent                    |
|-----------------------------------|----------------------------|
| <b>A. Lasing dyes</b>             |                            |
| Acridine Red                      | Ethanol                    |
| Coumarin                          | Water                      |
| 2,'7' Dichlorotfluorescein        | Ethanol                    |
| Esculin                           | Water                      |
| Fluorescein N <sub>a</sub> salt   | Water, Ethanol             |
| 7-Hydroxycoumarin                 | Water                      |
| $\beta$ Methylumbelliferone       | Water                      |
| Rhodamine 6G                      | Ethanol, Water, Alcohol    |
| Rhodamine B                       | Ethanol, Water             |
| Xanthene                          | Ethyl Alcohol              |
| <b>B. Dyes which did not lase</b> |                            |
| Acridine                          | Water+varying amounts acid |
| Acridine orange                   | Ethanol                    |
| Acridine yellow                   | Ethanol                    |
| Acridone                          | Ethanol Water              |
| Acriflavine                       | Ethanol                    |
| 9 Aminoacridine                   | Ethanol                    |
| Chloro aluminum phthalocyanine    | Ethanol                    |
| Dimethyl POPOP                    | Cyclohexane. thanol        |
| Eosin                             | Ethanol                    |
| Magdala red                       | Ethanol, Water             |
| Perylene                          | Cyclohexane, Ethanol       |
| Phenosafranine                    | Ethanol                    |
| Pyronine G                        | Ethanol                    |
| Rezazurin                         | Ethanol                    |
| Rubrene                           | Hexane                     |
| Safranine                         | Ethanol                    |
| Thionine                          | Ethanol                    |

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Although we know very little about the excited singlet absorption in most dyes, it is clearly shown in equation (6) and (19) that, if the excited singlet absorption cross section  $\sigma_a^*$  is large, lasing cannot be achieved, regardless of how strong the excitation is. There exist a number of dyes that have failed to achieve amplification by all means, although the fluorescence quantum efficiency is close to unity (one example is rubrene).

If all the parameters are known for a given dye, we can use the equation (19) to estimate the pump power for laser threshold or to calculate the gain for a given pump power. For instance, when  $n_{p0} = 2 \times 10^{23}$  photons/sec-cm<sup>2</sup>, the corresponding pumping intensity at wavelength 5000 Å is  $P_p/A = n_{p0}hc/\lambda_p = 8 \times 10^4$  watts/cm<sup>2</sup>. By measuring laser threshold under a variety of conditions, Webb et al.<sup>8</sup> determined the quantity  $K_{STT}$  for Rhodamine 6G in a methanol solution.

### III. EXPERIMENT\*

#### A. Construction of Dye Cell and Flashlamp

The design of a suitably fast excitation source was the central problem in the early dye laser experiments, because a rapidly rising excitation flash can overcome the effects of intersystem crossing that have been described in the theory. The first coaxial flashlamp excited dye laser, designed by Sorokin et al.<sup>3</sup>, utilized a novel coaxial flashlamp which discharged 100 joules in a pulse with a risetime to peak intensity of about  $0.3 \mu\text{s}$  and a duration of approximately  $1 \mu\text{s}$ . Our coaxial flashlamp is fashioned according to their general idea but in different design (see Fig. 3).

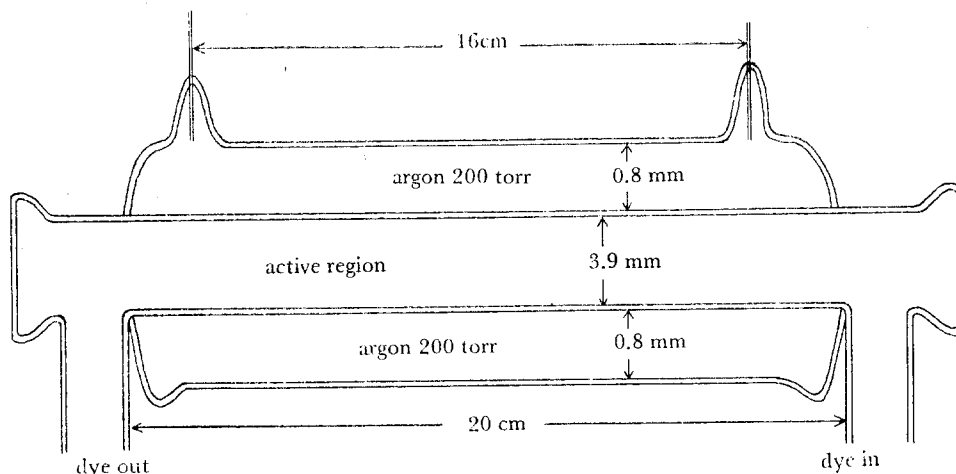
The laser cell is a 20-cm-long quartz tube with optical-quality glass windows mounted on the ends. This contains the organic dye which flows through cuvette to prevent heating, by replacing the dye solution between pulses. A second quartz tube surrounds the laser cell in coaxial fashion and serves as the external envelope of the lamp. The second tube is wrapped by the aluminium foil in order to reflect all flashlight back to dye cell. The discharge takes place between two tungsten thin rod electrodes placed 16 cm apart in the 0.8 mm thick annular space left between the two quartz tubes. This annular region was filled with about 200 torr argon which was used for gas-discharge medium. The diameter of the annular space is 5.5 mm. No advantage was found in the use of gases other than air in the lamp, and the experiments of Sorokin et al.<sup>13</sup> were conducted with an air-filled lamp which was fired by reducing the pressure in the discharge region.

The relatively large length of the inner tube (20 cm) exceeds that of the discharge region (16 cm); this sort of design permits easy installation of side stems for circulation of the dye solution through the inner tube. The circulative system has been done by a pump motor. The dye solution is kept constantly flowing while the laser is fired repeatedly. The refractive index of liquid solvent

\* This experiment was done at Northern Illinois University, DeKalb, Illinois, 60115 U.S.A.

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tends to vary quite strongly with temperature; hence accumulative distortions in optical quality of the laser are thus produced unless dye solution is circulated and there are no bubbles present in this dye solution.



**Fig. 3. Construction of Dye Cell and Flashlamp.**

### B. Circuitry of the Laser

The lamp can be flashed by a triggering scheme sketched in Fig. 4. The circuit consists of a  $0.3 \mu\text{f}$  cylindrical coaxial capacitor operated at 17.4 KV, a 20 KV DC power supply, a demountable spark gap, and a Tesla Coil. When the Tesla Coil ionizes the gas in the spark gap, the full voltage of the capacitor is applied across flashlamp terminals. During discharge, current flows from A, through the spark gap to B, then through the lamp. This flashlight can stimulate the dye molecules to the excited state. When the dye molecules return to the ground state ( $S_1 \rightarrow S_0$ ), the laser emission occurs. This phenomenon was recorded on photographic plate in a Jarrell Ash Spectrograph.

The five resistors, shown at right in the Fig. 4, function as bleeders, draining charge slowly from the capacitor. Without this safety provision the capacitor would retain a substantial portion of its lethal charge indefinitely. Do not touch the power supply circuit until the capacitor has been short-circuited, even though the circuit includes bleeder resistors. Short-circuiting can be done by briefly connecting a short length of copper wire across the terminals of the capacitor. The wire is fixed to a dry wooden handle about one meter long.

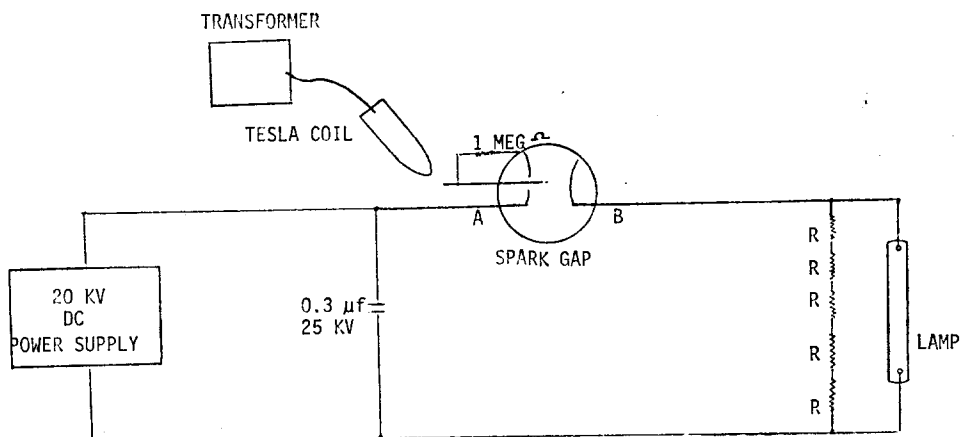


Fig. 4. Circuitry of the Laser.

### C. Performance

The experimental arrangement of the whole system, used for the dye laser studies, is sketched in Fig. 5. Dye cell is placed between one total reflection mirror and one 50% (output) transmission mirror. Both of them are at right angles to the spectrograph, so another total reflection mirror is set at 45 degrees to the dye cell in order to reflect completely the laser coming from the dye cell onto the spectrograph. The Jarrell Ash 3.4 meter grating spectrograph, with 1180 lines per millimeter grating, gives a dispersion of  $2.4 \text{ \AA}^\circ$  per millimeter in the first order.

It was found that the wavelength of Rhodamine 6G dye laser is  $5650 \text{ \AA}^\circ$  to  $6200 \text{ \AA}^\circ$ . From the "electrode to slit distance versus wavelength to set distance for best focus for wavelength being used" graph, the best distance for wavelength,  $5800 \text{ \AA}^\circ$  to  $6000 \text{ \AA}^\circ$ , being used is 52 cm, hence a spherical lens is positioned to focus the source to a point at 52 cm in front of the slit. However, the cylindrical lens is placed to obtain a vertical line on the slit of spectrograph. The counter reading is set at 2076 for  $6000 \text{ \AA}^\circ$ . During the alignment process a  $\text{H}_\alpha\text{-N}_\alpha$  laser was used to line up the optics. However, the optical alignment is always performed with the dye solution flowing. The alignment was rechecked before exposure of each plate.

A hollow iron cathode tube, producing a reference standard wavelength, is placed about 30 cm from and perpendicular to the optical axis. A plane mirror may be slid into position to reflect the iron light onto the slit. The exposure time for the iron spectrum was 4 minutes. The slit width required for the iron

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and laser spectrum was 60 microns. The height for the laser spectrum was 6 mm and 1 mm for the iron spectrum overlapping the laser spectrum. The hollow cathode tube was operated at 15 milliamperes which is slightly below the rated current in order to protect the tube.

A cylindrical coaxial  $0.3 \mu\text{f}$  capacitor was used for stored electrical input energy and discharge energy; this capacitor is connected to a high voltage DC power supply. With the Rhodamine dyes the best conversion efficiency attained was about  $0.4\%$ <sup>13</sup>. For 50 joules of capacitively stored electrical input energy ( $0.3 \mu\text{f}$  at 18.2 Kv) the laser typically produced an output pulse whose peak power was about 1 MW. The total output energy contained in the pulse is about 0.2 joules. If the actual shape of the light pulse may be considered as a triangle with the basewidth of 400 nsecond, the rise-time to the peak pulse then works out to be about 200 nsecond.

The Kodak Spectrum Analysis Plates 103F suitable for wavelength  $4000 \text{ \AA}$  to  $7000 \text{ \AA}$  were used. After several spectra being taken, the plates were developed in Kodak D-19 solution for 4 minutes, rinsed 30 seconds in  $20 \text{ }^\circ\text{C}$  water, and then fixed in Kodak fixer for 10 to 15 minutes. After fixing, the plates were washed in running  $20 \text{ }^\circ\text{C}$  water for 30 minutes. Several traces of spectrum were made on the plates. Each exposure consists of laser spectrum overlapped by an iron spectrum. Comparing the iron spectrum to the known standard iron spectrum, the wavelength of the dye laser can be found very easily.

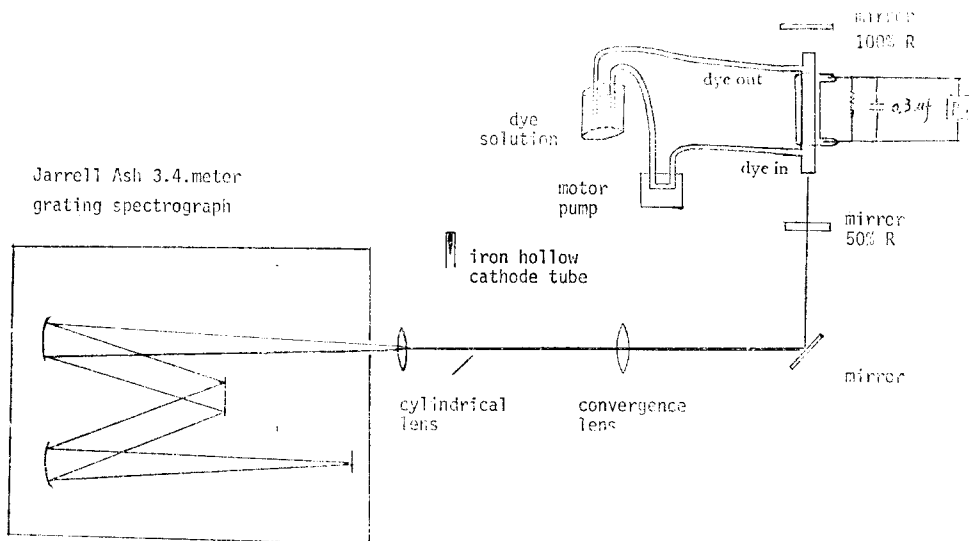
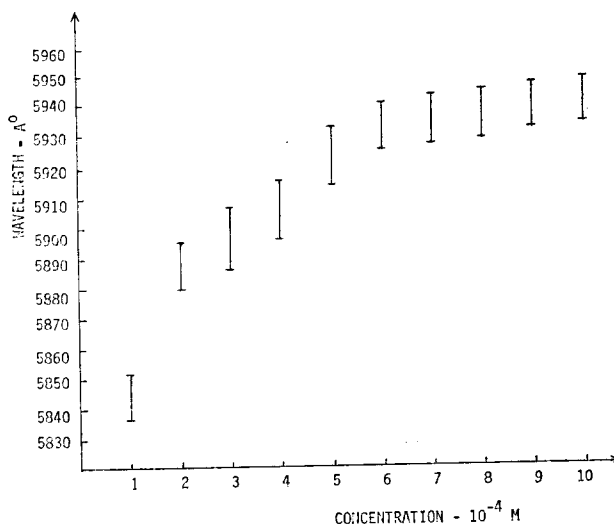


Fig. 5. The Experimental Arrangement of the Whole System.



#### IV. RESULTS AND CONCLUSION

Rhodamine 6G has high fluorescence quantum yield. We have found that  $10^{-4}$  M solution of Rhodamine 6G in alcohol is very nearly an optimal concentration for obtaining reliable, low threshold, and reasonably energetic laser action. From the experimental results, shown in Fig. 6, it was found that the lasing wavelength increasing as the concentration of dye solution being increased. In other words, the lasing frequency will decrease as the concentration of the dye solution is increased. This is the same effect that was discussed at length in Ref. 3. Moreover, comparing our results to the previous investigator's (see table 2), it seems close to each other. Generally speaking, the lasing wavelength of dye laser also depends on length of dye cell and geometry of cavity, but it will be in a range of wavelength.



**Fig. 6. The Relationship Between Wavelength and Dye Concentration.**

As previously discussed, laser action takes place when the gain is a maximum and the lasing frequency at threshold. This situation depends on the population of dye molecules in the excited singlet state. Hence, the concentration of molecules in the excited singlet state  $S_1$  must reach a certain value, the critical inversion, before coherent emission will be reproduced by the dye. The magnitude

of the critical inversion depends upon the losses of the complete laser system. The resultant high optical loss is a serious impediment to laser action.

**Table 2. Summary of Lasing Wavelength of Rhodamine 6G**

| Solvent      | Molar Concentration   | Wavelength A° | Reference    |
|--------------|-----------------------|---------------|--------------|
| Alcohol      | $1 \times 10^{-4}$ M  | 5838 - 5852   | Present Work |
| Alcohol      | $2 \times 10^{-4}$ M  | 5882 - 5906   | Present Work |
| Alcohol      | $4 \times 10^{-4}$ M  | 5906 - 5918   | Present Work |
| Alcohol      | $5 \times 10^{-4}$ M  | 5912 - 5934   | Present Work |
| Alcohol      | $6 \times 10^{-4}$ M  | 5924 - 5940   | Present Work |
| Alcohol      | $7 \times 10^{-4}$ M  | 5928 - 5942   | Present Work |
| Alcohol      | $8 \times 10^{-4}$ M  | 5930 - 5946   | Present Work |
| Alcohol      | $9 \times 10^{-4}$ M  | 5934 - 5948   | Present Work |
| Alcohol      | $10 \times 10^{-4}$ M | 5936 - 5950   | Present Work |
| Ethanol      | $1 \times 10^{-4}$ M  | 5790          | 12           |
| Ethl Alcohol | $1 \times 10^{-4}$ M  | 5850          | 3            |
| Ethanol      | $2 \times 10^{-4}$ M  | 5900          | 14           |
| Ethanol      | $1 \times 10^{-4}$ M  | 5950          | 15           |
| Ethanol      | $5 \times 10^{-4}$ M  | 5950          | 18           |
| Ethanol      | $2 \times 10^{-4}$ M  | 5990          | 19           |

Obviously, intersystem crossing competes with fluorescence in the deactivation of  $S_1$ . This competition is detrimental to the operation of the laser in several ways. Owing to the relatively long life the triplet state can thus act as a trap for the excited molecules and deplete the supply of molecules available for the laser process. Hence, steady-state lasing is prevented by triplet state effects. Simultaneously, there have been some singlet losses as well. However, from equation (19), it was found that the sufficiency strong excitation can overcome the singlet losses. Especially, the shorter the fluorescence lifetime is, the higher excitation is required, because the product of the lifetime and pumping intensity should be larger than threshold for the laser action.

Because of the continual buildup of triplet population, it is important that the optical pumping for dye laser must have a sufficiently short risetime to reach threshold gain before triplet losses become dominant. The required pumping conditions are dependent, in a complicated way, upon characteristics of both flashlamp and dye properties. For the purpose of reducing the lasing threshold, it is effective to make the flashlamp and the dye cell small, and to get the density of pumping light in the cell to increase. The low threshold value is caused not only by the fast risetime but also by the small diameter of the cell, because the threshold value is almost independent of the increase of risetime up to 50 nsecond.

Laser emission occurs only if the dye is exposed to white light of sufficient intensity. The intensity of the light varies inversely with the rate of discharge. The capacitor should be specially designed to discharge in a very brief time interval, because a certain number of molecules will accumulate in the lowest triplet state, a metastable level, during the time required for the pumping flux to reach its peak. Hence, ordinary capacitors discharge more slowly and will not work in this application.

The flashlamp has proved to be extremely reliable, output energy and flash profiles being completely reproducible. In order to increase the output power without destroying the lamp, the duration of the pulse and the energy of the laser, which even for the best of the Rhodamine 6G dyes is not usually more than a few joules, practical application, however, often calls for much greater output energies.

## APPENDIX A

A good dye for beginning experiment is rhodamine 6G. This orange dye emits light that spans a spectral range of some 440 angstroms from yellowish-green to red. The molecular weight of the dye is 449. A  $10^{-4}$  molar concentration can be achieved by dissolving 0.045 gram of the dye in alcohol to make one liter. Similarly, a  $10^{-2}$  molar concentration can be gotten by dissolving 4.5 gram of the dye in one liter alcohol.

## ACKNOWLEDGMENTS

The author wishes to express his gratitude to Dr. Peter Tsao for excellent guidance, valuable suggestions throughout this research, helpful assisting in the experiment, and careful reading of the manuscript, and to Dr. Mazhar Hasan for many enlightening discussions and suggestions concerning a number of peculiarities that appeared in the theory, and to Dr. John C. Shaffer for his moral support during my stay at Northern Illinois University.

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