THE WAVELENGTH SPECTRUM SHIFT OF A CAVITY-DUMPED ARGON LASER-PUMPED RHODAMINE 6-G ORGANIC-DYE LASER

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THESIS

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by

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ABSTRACT

The wavelength spectrum of a CW pumped Rhodamine 6-G organic dye laser is observed to shift approximately 150 Angstroms toward shorter wavelengths when pumped by 30 nsec pulses at a one megahertz repetition rate from a cavity-dumped Argon laser. Experimental evidence of the shift is presented, gain and rate equations are developed for a simplified dye laser model, and theoretical results are obtained for a computer simulation of the experiment. A comparison is made of the theoretical and experimental results and satisfactory agreement is obtained within the limits of the values of the parameters used and the assumptions made in formulating the model.

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I. INTRODUCTION

Man's quest for knowledge, inherent curiosity, and need for a tunable coherent source of light have rapidly advanced the development of the organic dye laser since stimulated emission was observed from Trivalent Uranium (a four level laser system) by Sorokin and Stevenson [1] in 1960.

Brock, et al. [2] in 1961 proposed that organic compounds could be used as a laser medium, but stimulated emission from an organic material was not observed until 1962 when Morantz, White and Wright [3] observed it from benzophenone and napthalene imbedded in a glass matrix. This was followed in 1963 by the observation of stimulated processes from Eu-benzoylacetonate by Lempicki and Samuelson [4].

Stimulated emission from an organic dye, however, was not accomplished until 1966 when it was observed by Sorokin and Lankard [5] from Chloro-aluminum phthalocyanine dissolved in ethyl alcohol when pumped by a giant pulse ruby laser. Sorokin and Lankard [6] also reported in 1967 the first observed stimulated emission from flashlamp pumped Acridine Red, Rhodamine 6-G (the most common dye used in present day organic dye lasers), and Fluorescein. Schafer, Schmidt, and Volze [7] reported stimulated emission from several other organic dyes during this same time period while using the same basic procedure as Sorokin and Lankard [5], and they were the first to suggest that a dye laser operated on transitions from the excited singlet state to vibrational levels of the ground singlet state.

The feasibility of a continuous wave dye laser was discussed by Snavely [8] and was demonstrated by Snavely and Schafer [9] in 1969. CW operation was achieved in 1970 by Peterson, Tuccio and Snavely [10] and

Hercher and Pike [11] using an Argon ion laser as the pump source and Rhodamine 6-G as the organic dye.

The first CW dye lasers were only about one percent efficient. This has been raised to the present day level of thirty-five percent through the efforts of several groups of people including Hercher and Pike [12, 13], Kohn, Shank, Ippen and Dienes [14, 15], and Tuccio and Strome [16, 17].

The evolution of the dye laser has seen it constructed in many ways. The tunable distributed-feedback dye laser built by Shank, Bjorkholm and Kogelnik [18], the prism-dye laser constructed by Chandra, Takeuchi, and Hartmann [19], and the evanescent-field-pumped dye laser demonstrated by Ippen and Shank [20] are a few of the many sophisticated methods that have been used to obtain dye laser emission. The most common dye laser configuration, however, has the pump and dye laser cavities aligned with the beams coincident and the dye flowing perpendicular to the pump beam through a dye cell.

Several uses have been proposed for dye lasers. Bloom [21] has suggested its use for attenuation measurements on narrow-band absorption lines in molecular gases, and Sorokin, Lankard, Moruzzi and Hammond [22] have proposed using the dye laser for optical studies of rare-earth ions, photochemistry, and double-quantum absorption spectroscopy. The dye laser characteristics which make it exceptionally well suited to the above uses are its tunable range which can be as much as 1100 Angstroms from a single dye, and its narrow linewidth which is normally less than 0.5 Angstroms.

Uses for the dye laser which have already been published include infrared difference-frequency generation [23], megawatt tunable second harmonic and sum frequency generation [24], studies of the Sodium D

resonance lines by high resolution Spectroscopy [25], and detection of OH in the atmosphere [26]. Further uses of the dye laser are limited only by the interest and ingenuity of man.

In this work it has been observed that the output wavelength spectrum of a CW pumped Rhodamine 6-G organic dye laser shifts toward shorter wavelengths when pumped by short intense pulses from a cavity-dumped Argon laser. This shift to the green region of the visible wavelength spectrum is important in that the new spectrum can be matched to the wavelength response curve of a crystal to produce a tunable dye laser optical memory system. The new spectrum also provides the opportunity to investigate laser propagation in blue and green sea water over an appreciable wavelength range using a single dye.

The purpose of this thesis is to provide experimental results of the observed spectrum shift, to obtain a computer simulation of a dye laser model, and to compare experimental and theoretical results. Section II discusses dye laser theory based on energy level considerations and gives the development of the dye laser gain and rate equations for an assumed physical model. Section III presents experimental procedures and results and Section IV discusses the solution procedures and results of the theoretical model. Section V provides a comparison of experimental and theoretical results and conclusions. The computer program used to solve the dye laser model is listed in Appendix A.

II. DYE LASER THEORY

A. ORGANIC DYE PROPERTIES

1. Energy Level Description

A typical energy level diagram for an organic dye is shown in Figure 1. The reference level A is the electronic ground state of the molecular singlet state S_0. S_1 and S_2 are the first and second excited singlet states, and T_1 and T_2 are the first and second excited triplet states. The small letters a and b indicate molecular vibrational energy levels and the primed letters indicate molecular rotational energy levels within a state. The typical separation of vibrational energy levels is 0.1 electron volts and that of rotational energy levels is 0.001 electron volts [27] so that each state may be viewed as a continuous band of energy.

2. Dye Laser Process

The dye laser process starts with the excitation of molecules from level A of S_0 to an upper vibrational or rotational level of S_1. The excited molecules then decay very rapidly (10^{-10} -10^{-13} sec) [28] by nonradiative internal conversion to B.

The excited molecule in B has three options. It may decay spontaneously to a or a' (called fluorescence), it may undergo a stimulated transition to a or a', or it may travel via intersystem crossing to the lower level of T_1.

Fluorescence depends on the natural lifetime of B. The fluorescence spectrum of an organic dye is governed by the Franck-Condon principle [29] which states that preferred electron transitions are determined by the wave functions of the individual energy levels.

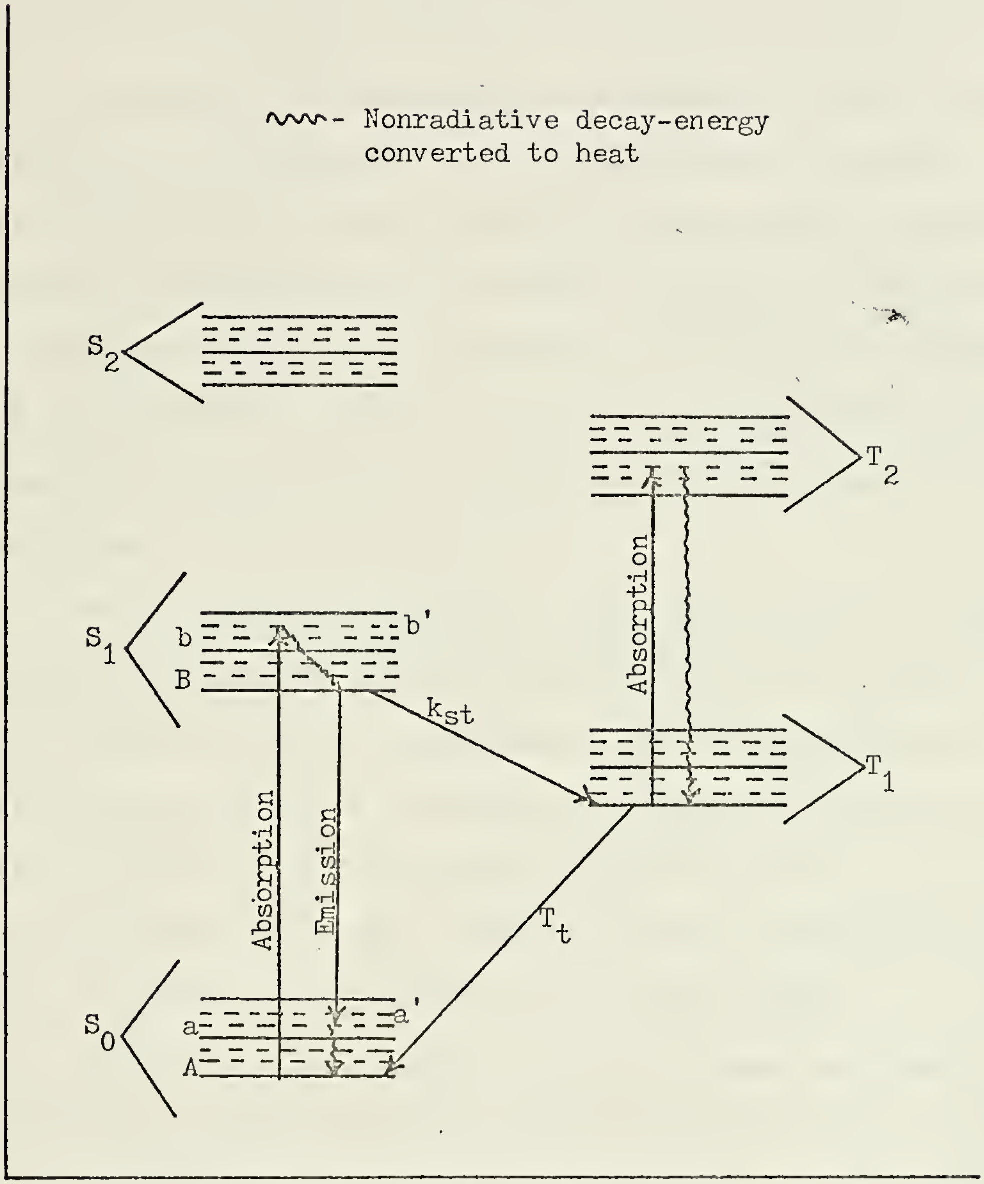


Figure 1: Organic dye energy levels.

Stimulated transition to a or a' can occur only if the excitation pulse is fast and intense enough to create a much larger than normal concentration of molecules in S_1, called critical inversion, so that coherent emission may take place from the dye. The critical inversion as well as the emission wavelength are governed by losses in the dye laser system. Once the excited molecule reaches a or a' it decays nonradiatively to A.

Intersystem crossing is the least desirable transition for an excited molecule. The singlet state is characterized by opposite electron spins and the triplet state by parallel electron spins. As a result, according to the laws of quantum mechanics, the $S_1 - T_1$ transition is spin forbidden and is in fact approximately 10^{-6} [30] less probable than the $S_1 - S_0$ transition. However, the $S_1 - T_1$ transition does occur and is significant enough to have an important negative effect on the dye laser process.

The intersystem crossing rate time constant, k_{st}^{-1}, is generally much smaller than the T_1 state lifetime, T_t, therefore, state T_1 acts as a time dependent trap for dye molecules. This not only reduces the possible dye laser efficiency by removing dye molecules from the singlet system, but also, since the absorption spectrum of the $T_1 - T_2$ transition of an organic dye usually overlaps the dye fluorescence spectrum, another loss mechanism is added to the dye laser system. $S_1 - S_2$ absorption is possible but it is usually neglected because the fraction of molecules in S_1 needed to achieve critical inversion is very small [31].

Transitions to A are generally assumed to be from the lowest level of T_1. These transitions may be either radiative, called phosphorescence, or, as is the situation for most organic dyes, nonradiative. The longer T_t is, the faster the dye pumping pulse has to be to excite

enough molecules to reach critical inversion before the T_1 trap prevents dye laser emission.

The T_1 trap problem has caused a lot of research to be done to find substances which will shorten T_t via collisions with molecules in T_1. This action is called quenching and two very effective quenching agents are cyclooctatetraene (COT) and molecular oxygen.

B. DYE LASER MATHEMATICS

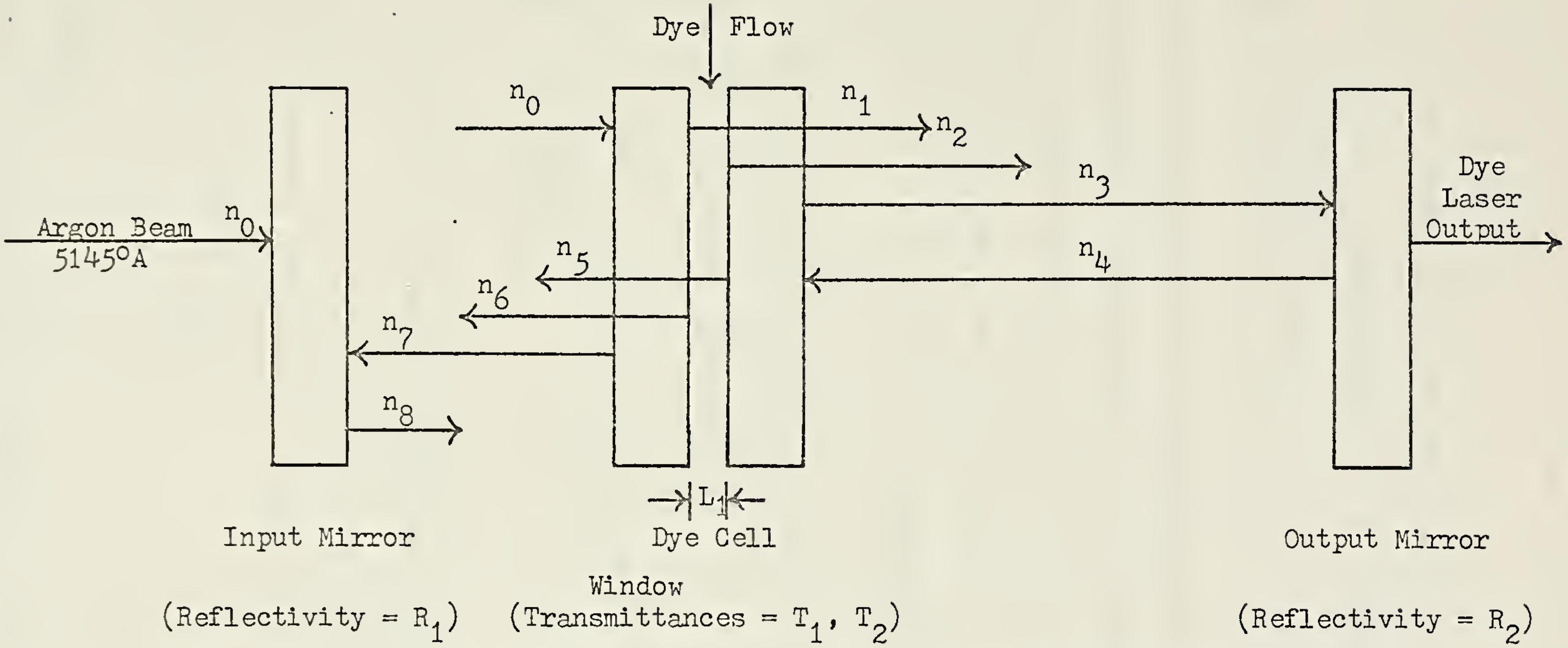
1. Dye Laser Gain Equation

The dye laser gain equation is a mathematical statement of the production and loss rates for photons in the laser cavity. Photons are produced by stimulated emission from the excited singlet state to the ground singlet state. Photons are lost by S_0 - S_1 and T_1 - T_2 absorption and by loss through extrinsic means such as output mirrors, optical surface scattering, etc. Photon production is also possible by stimulated emission from the excited triplet state but nonradiative decay is so rapid from this state to the lower triplet state that this source of photons can be neglected.

A simplified schematic diagram of a dye laser is shown in Figure 2. L_1 is the dye cell length, R_1 and R_2 are the input and output mirror reflectivities, T_1 and T_2 are the dye cell window transmittances, and n_0, n_1, n_2, etc. are the photon densities at any time at the indicated positions of a round trip in the laser cavity. This particular cavity is used as the model for the following derivation of the gain equation.

The rate of photon production per unit length in the dye may be written as follows:

$$\frac{dn}{dl}_{\text{total}} = \frac{dn}{dl}_{\text{emission}} - \frac{dn}{dl}_{\text{absorption}} \tag{1}$$



n_0, n_1, n_2, etc. = number of photons/cm^3 at any time during a round trip in the laser cavity

Figure 2: Dye laser cavity schematic.

The first term on the right hand side of (1) is photon production by stimulated emission and the second term is photon loss by singlet and triplet state absorption.

Yariv and Gordon [32] have shown that the expression for the transition rate caused by a monochromatic beam of light of wavelength λ is

$$\frac{dn}{dt}_e = \frac{N_s \lambda^4 E(\lambda) n(\lambda)}{8 \pi T_s \bar{n}^3} \text{ cm}^{-3} \text{ sec}^{-1} \tag{2}$$

where N_s is the population density of the excited singlet state, $n(\lambda)$ is the number of photons of wavelength λ per cm^3 of active medium, T_s is the excited singlet state lifetime, $\bar{n}$ is the index of refraction, and $E(\lambda)$ is the fluorescence lineshape function normalized so that

$$\int_0^\infty E(\lambda)\, d\lambda = \emptyset = \text{quantum yield} = \frac{\text{photons emitted}}{\text{photons absorbed}}.$$

An expression for the first term of (1) can now be derived. Since

$$\frac{dn}{dt}_e = \frac{dn}{dl}_e \times \frac{dl}{dt} \quad \text{and} \quad \frac{dl}{dt} = \text{velocity} = \frac{c}{\bar{n}}$$

where c is the speed of light, it follows that

$$\frac{dn}{dl}_e = \frac{N_s \lambda^4 E(\lambda) n(\lambda)}{8 \pi c T_s \bar{n}^2} \quad . \tag{3}$$

The molecular extinction coefficient, which is the absorption cross-section for a single dye molecule, can be used to write the singlet and triplet absorption loss per unit length for (1). These terms are

$$\frac{dn}{dl}_{singlet} = n(\lambda)\, N_o\, \sigma_s(\lambda) \tag{4}$$

$$\frac{dn}{dl}_{triplet} = n(\lambda)\, N_t\, \sigma_t(\lambda) \tag{5}$$

where N_o and N_t are the population densities of the ground singlet and triplet states, and σ_s and σ_t are the wavelength dependent singlet and triplet absorption cross-sections.

Let

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

and substitute (3), (4) and (5) into (1). This yields the total production rate of photons per unit length in the dye medium which is

$$\frac{dn}{dl}_{total} = n(\lambda)\ \sigma_e(\lambda)N_s - n(\lambda)\ \sigma_s(\lambda)N_o - n(\lambda)\ \sigma_t(\lambda)N_t. \qquad (6)$$

Separation of variables of (6) and integration over the length of the dye cell with the boundary conditions that where $l = 0$, $n = n_1$, and where $l = L_1$, $n = n_2$, yields the solution

$$n_2 = n_1 \exp[\sigma_e N_s - \sigma_s N_o - \sigma_t N_t]L_1. \qquad (7)$$

The n's and σ's are still wavelength dependent and from Figure 2 it can be seen that since the paths traveled by photons n_1 and n_5 are through the same medium but in opposite directions that (7) also holds for the relation between n_6 and n_5.

It is obvious from Figure 2 that $n_1 = n_0T_1$, $n_3 = n_2T_2$, $n_4 = n_3R_2$, $n_5 = n_4T_2$, $n_7 = n_6T_1$, and $n_8 = n_7R_1$. Substitution of these relations consecutively into (7) gives the result

$$n_8 = n_0\exp[\sigma_eN_s - \sigma_sN_o - \sigma_tN_t + \frac{1}{2L_1}\ln(R_1R_2T_1{}^2T_2{}^2)]2L_1. \qquad (8)$$

Let $G(\lambda)$ be the equation in brackets in (8) and define it to be the gain. Then

$$n_8 = n_0\exp[2\ G(\lambda)\ L_1], \text{ and}$$

$$G(\lambda) = \sigma_eN_s - \sigma_sN_o - \sigma_tN_t + \frac{1}{2L_1}\ln[R_1R_2T_1{}^2T_2{}^2] \qquad (9)$$

which is the desired gain equation and is similar to the results obtained by Snavely [8] and McColgin, et al. [33].

2. Dye Laser Rate Equations

The dye laser will lase at the value of λ where (9) is a maximum. To find this wavelength it is necessary to solve the state population density rate equations for the dye laser. These equations are

$$\frac{dN_s}{dt} = -\frac{1}{T_s}N_s + P(t)N_o \qquad (10A)$$

$$\frac{dN_t}{dt} = -\frac{1}{T_t}N_t + k_{st}N_s \qquad (10B)$$

$$\frac{dN_o}{dt} = -P(t)N_o + \left(\frac{1}{T_s} - k_{st}\right)N_s + \frac{1}{T_t}N_t \qquad (10C)$$

$$N = N_o + N_s + N_t \qquad (10D)$$

N is the total population density and (10D) is true only for an enclosed system undergoing no photochemical processes. N_t, N_o, and N_s are as defined for the gain equation, T_s and T_t are the lifetimes of the singlet and triplet states, k_{st} is the excited singlet to ground triplet intersystem crossing rate constant, and P(t) is the optical pumping rate.

There are several assumptions made in writing the rate equations in the form of (10A), (10B), (10C) and (10D). These assumptions are:

(a) The number of molecules involved in intersystem crossing from the triplet to the singlet state is negligible.

(b) The molecules initially excited into the first excited singlet state reach thermal equilibrium in a short time compared to the pumping time.

(c) Ground triplet to excited triplet state absorption is not negligible, but the population of the excited triplet state is negligible because of rapid nonradiative decay.

(d) The dye laser has essentially only three levels. (The fine vibrational and rotational levels in each main state are neglected.)

(e) The effect on state population densities caused by dye self-absorption of the lasing wavelength is negligible. (There are approximately two orders of magnitude difference between the absorption coefficient for the pumping wavelength and the expected lasing wavelength so this assumption is not too restrictive.)

(f) The point of maximum gain is reached so fast that the effect of reduction of the excited singlet state due to stimulated emission can be neglected. (This assumption has been shown to be realistic by Atkinson and Pace [34] and is very true at the onset of lasing; however, as lasing progresses and photon buildup occurs in the laser cavity the stimulated emission term can become very significant. The same is true of the self-absorption term since it also depends on intracavity power.)

Equations (9), (10A), (10B), and (10D) are used to evaluate the dye laser model in a computer simulation in Section IV. Equation (9), the gain equation, is used in the discussion of theoretical results to explain the theoretical time dependent wavelength sweep of the dye laser.

III. EXPERIMENTAL PROCEDURES AND RESULTS

A. EXPERIMENTAL SET UP

The apparatus used in generating and measuring the output of the Argon and dye lasers was set up as shown in Figure 3. A Beck Wavelength Reversion Spectroscope with a single line resolution of approximately four Angstroms was also used to monitor the dye laser output wavelength.

The beam splitter indicated in Figure 3 was actually an integral part of the front end of the Argon laser and is normally used as part of a power monitoring system. This system was modified so that the Argon laser output pulse could be monitored on an oscilloscope via a photodiode detector.

The diode detector and power meter were both tested for linearity and saturation at higher powers than could be expected at their locations in Figure 3. They both exhibited no signs of approaching saturation and their deviations from linearity over the testing range were minimal.

The risetime of the Argon laser is normally limited to seven nanoseconds because of cable transit times, etc., however, in the particular set up of Figure 3 the risetime was limited to 12 to 15 nanoseconds because that was the lower limit of the pulse generator risetime. An explanation of the cavity dump mechanism can be found in Maydan [35].

B. EXPERIMENTAL PROCEDURES

The first step in the procedure was the alignment of the Argon laser cavity dump system. This system consisted of a curved front half-mirror with vertical and horizontal adjustments, a rear curved mirror with vertical and horizontal plane tilt controls, and an acoustooptic cell housing

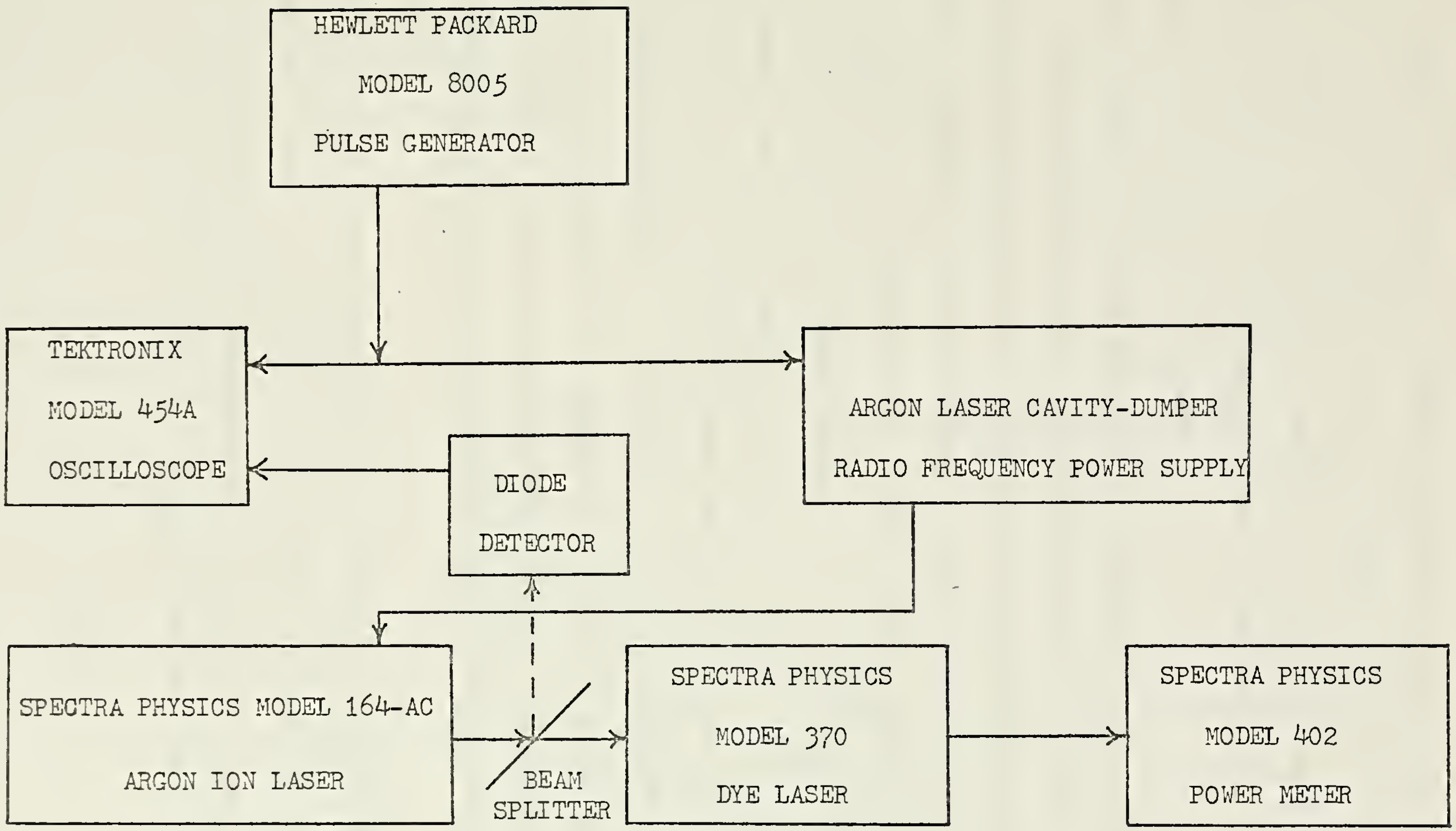


Figure 3: Experimental set up.

with controls to adjust its longitudinal and vertical positions as well as its angular alignment. All of these controls had to be precisely set for the cavity dump system to function properly, and the Argon laser power output and pulse shape had to be constantly monitored because some of the adjustments were extremely delicate. A slight movement of the rear mirror adjustments, for example, severely degraded the Argon laser output pulse and beam mode structure.

The next step in the procedure was the alignment of the Argon and dye lasers. This was done on a specially designed platform which had permanent hold down clamps for the Argon laser and kinematic mounts for the dye laser. Alignment of the two lasers was very sensitive and was a critical part of the experimentation since any nonparallelism of the dye and Argon laser cavities severely reduced the power conversion efficiency.

The Argon laser power supply was set for a current of 30 amperes after alignment was achieved. With the pulse generator delivering three volt, thirty nanosecond pulses at a repetition rate of one megahertz to the cavity-dump rf power supply, the average power output of the Argon laser was 500 milliwatts. These values and settings were then used as Argon laser operating reference points and standards for the rest of the experimentation.

The final step in the procedure was to monitor the output of the Argon and dye lasers. The Argon and dye laser powers were recorded, the Argon laser pulse shape was sketched, and the various dye laser wavelengths noted as the pulse width of the pulse generator was varied.

C. EXPERIMENTAL RESULTS

The results yielded some new and interesting properties of the dye laser. The wavelength of peak output power and efficiency of the cavity-dumped laser-pumped dye laser was found to occur at a shorter wavelength than that of the continuous wave laser-pumped dye laser. In addition, the tuning range of the pulsed dye laser was narrowed.

The normalized output for both the continuous-wave laser pumped and pulsed-laser pumped dye laser are shown for comparison in Figure 4. The same dye laser was used to obtain both curves.

A two piece mathematical model which is an excellent fit to the actual Argon laser output pulse used to obtain the pulsed dye laser curve is shown in Figure 5. The repetition rate of this pulse was one megahertz.

It was also discovered that as the pulse width driving the cavity-dumped rf power supply was lengthened the wavelength of peak emission shifted toward longer wavelength and the laser efficiency decreased. In addition, for a given pulse width, an increase in the output power of the Argon laser (accomplished by increasing the current to the Argon laser power supply) shifted the peak emission wavelength to shorter wavelengths.

The lowest peak wavelength observed was 5650 Angstroms and the highest conversion efficiency obtained was almost thirty percent. This same dye laser pumped with a continuous wave laser had an efficiency of only seventeen percent at the peak output wavelength of 5800 Angstroms.

The fluorescence spectrum of the dye in the laser was also measured by use of a spectroscope with a high intensity white light source. This spectrum along with the wavelength-power curve of the cavity-dumped laser-pumped dye laser are compared in Figure 6. It is obvious from this figure that the peak lasing wavelength for the pulsed pump is approaching the peak of the dye fluorescence curve.

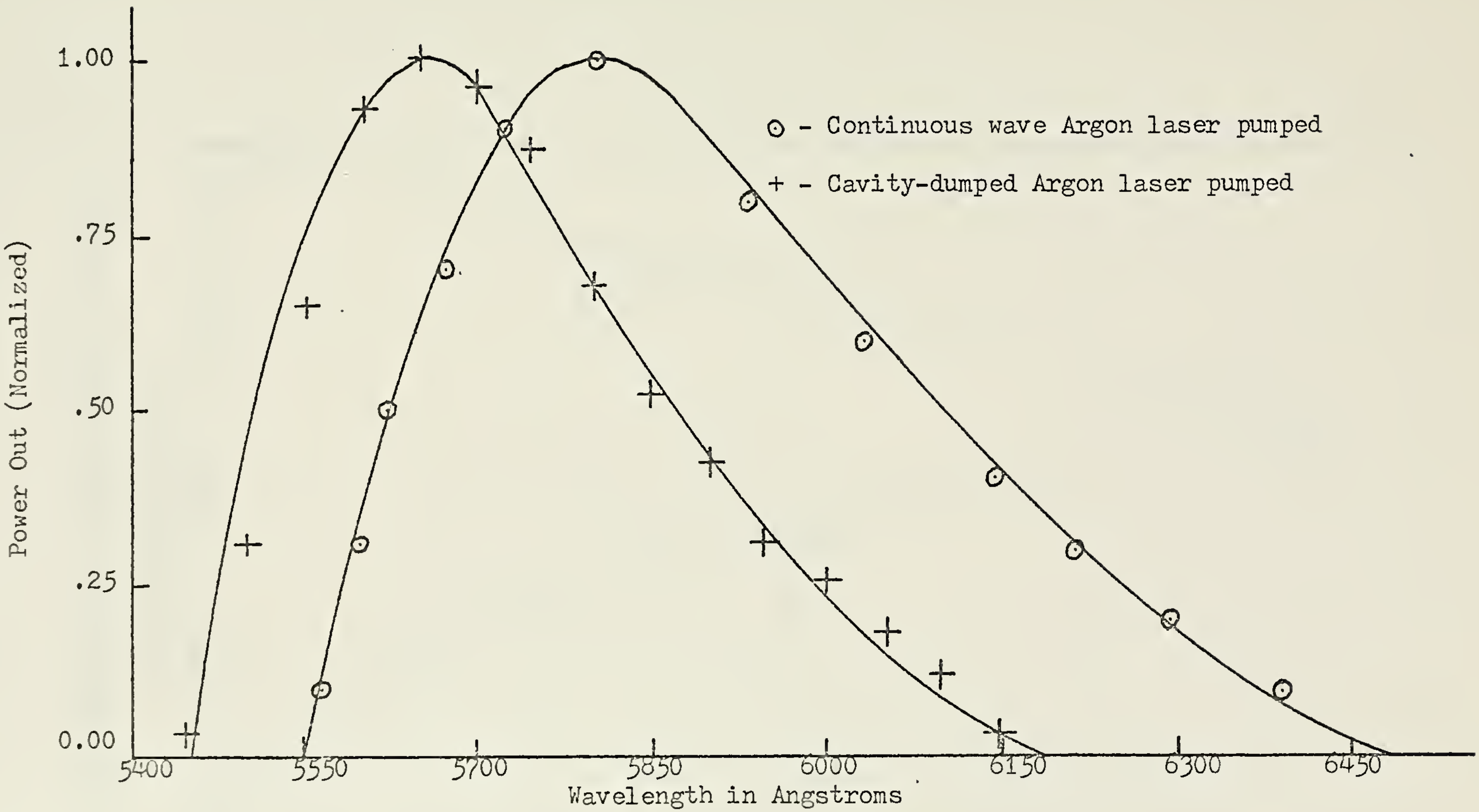


Figure 4: Power - wavelength spectrums of a Rhodamine 6-G dye laser.

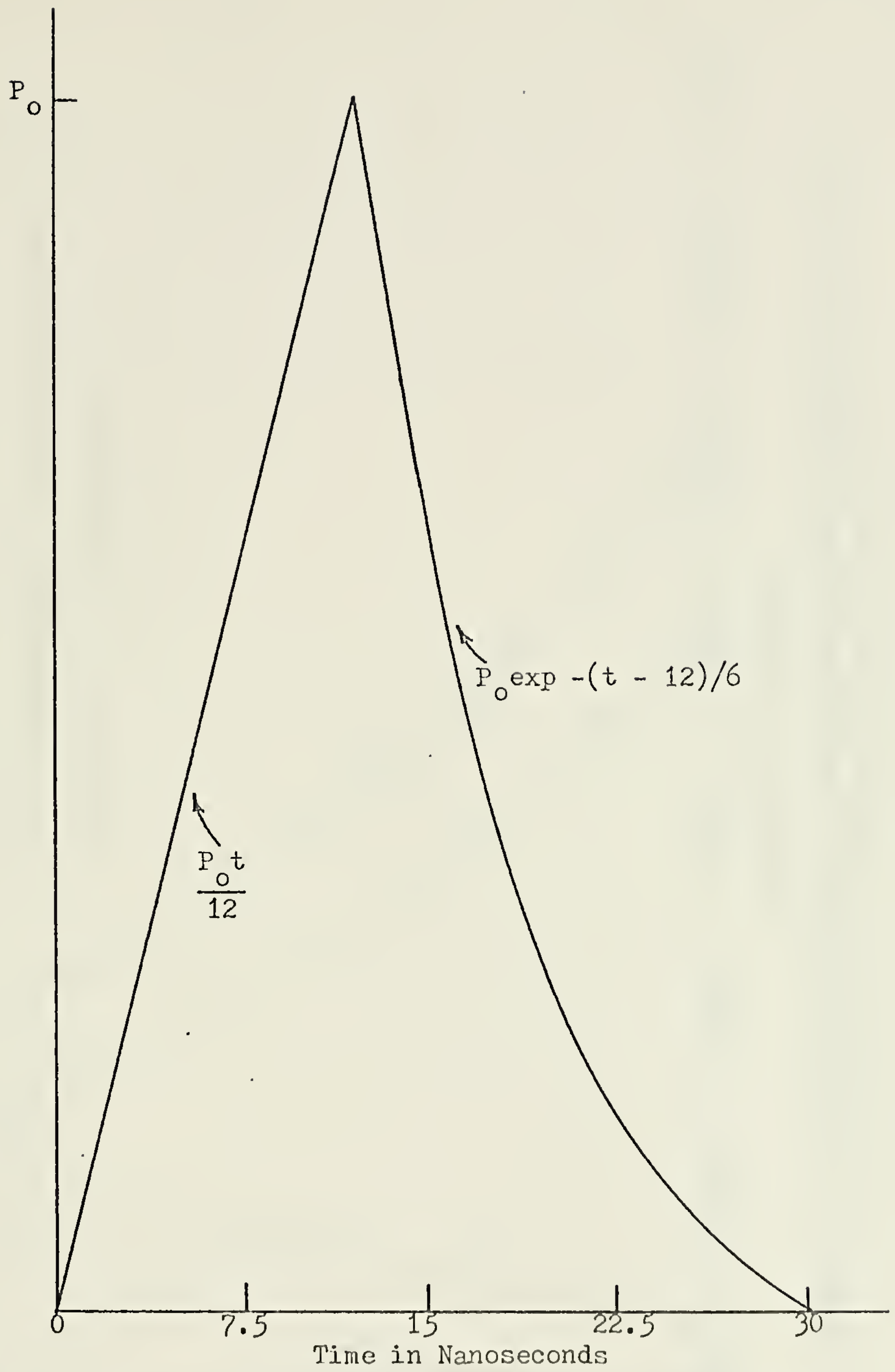


Figure 5: Typical Argon laser output pulse.

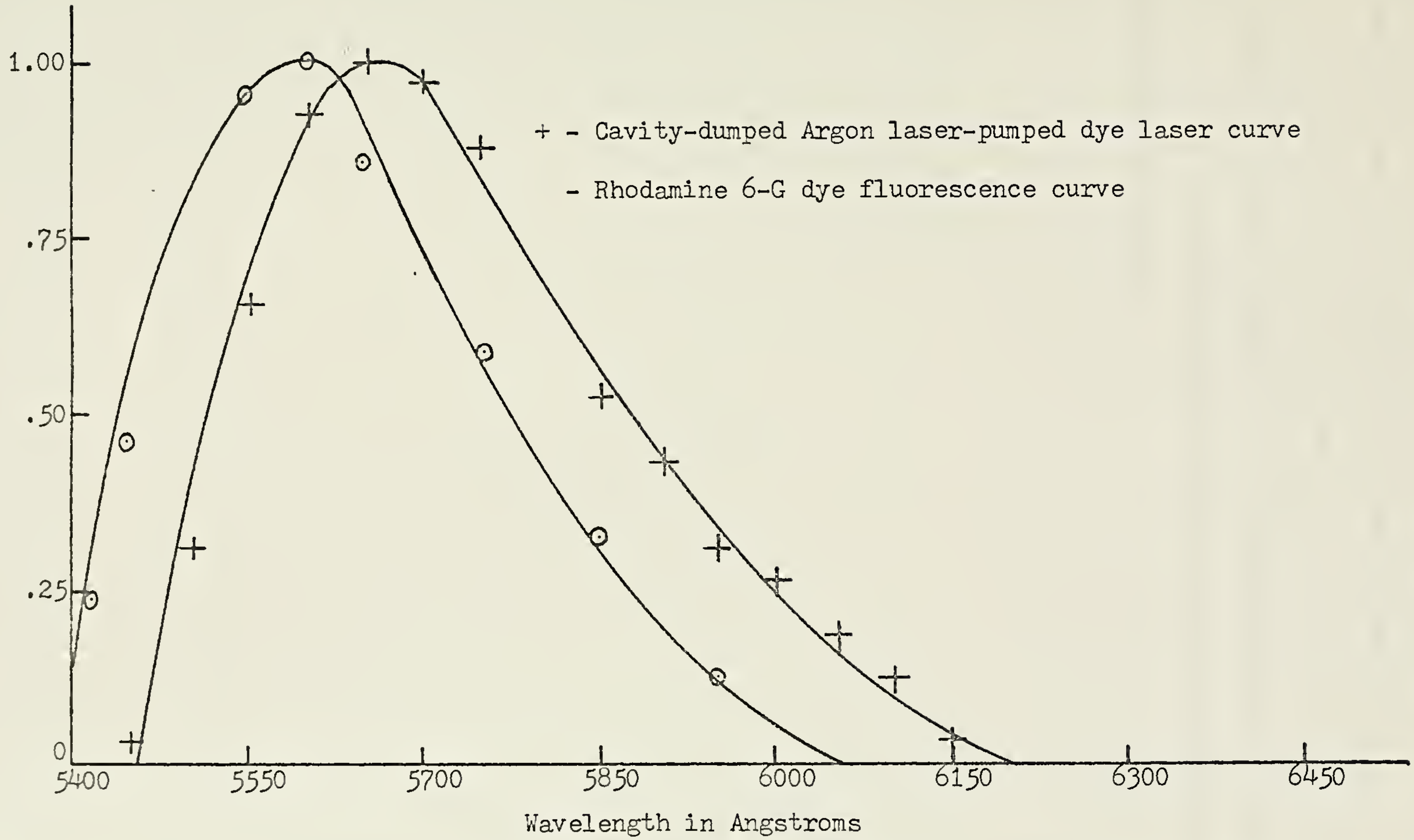


Figure 6: Wavelength-power curve of cavity-dumped Argon laser-pumped dye laser and Rhodamine 6-G fluorescence curve (Normalized).

A summary of the significant experimental observations is as follows:

(1) The pulsed dye laser wavelength spectrum exhibits a substantial green shift from the spectrum of the CW pumped dye laser.

(2) The shift increases and approaches the dye fluorescence curve as the pulse length from the cavity-dumper gets shorter.

(3) The efficiency is greater and the tuning range shorter for the pulsed dye laser.

IV. DYE LASER MODEL EVALUATION

Experimental results showed that the output wavelength spectrum of the cavity-dumped laser-pumped dye laser approached the dye fluorescence curve. The dye laser model of Figure 2 is analyzed in this section using equations (9), (10A), (10B) and (10D), which are the model gain and rate equations, and the pumping waveform of Figure 5 to determine if the spectrum shift can be predicted.

A. LITERATURE SURVEY

The first application of applying rate and gain equations to the dye laser was made by Sorokin, et al. [36, 22]. Their application assumed a Gaussian waveform pumping pulse and they attempted to predict the transient pulse shape of a pulsed Chloro-aluminum phthalocyanine (CAP) dye laser. They concluded from their results that a rapid pulse risetime was necessary or triplet state accumulation could prevent dye laser action.

The second analysis of Sorokin and his group again assumed a Gaussian pumping pulse and attempted to predict the efficiency of various dye lasers as functions of quantum efficiency and the excited singlet to triplet intersystem crossing rate constant (k_{st}). The results from this work yielded the prediction that dye laser efficiency should increase as the intersystem crossing rate time constant decreased or as the quantum efficiency increased.

A year later, in 1968, Bass, Deutsch and Weber [37] used the rate and gain equations to predict lasing frequencies and time to lasing for Gaussian shaped laser and flashlamp pumped dye lasers. They had

experimentally observed that a laser pumped dye laser emitted a shorter wavelength than when flashlamp-pumped, however, different laser cavities were used for the two pumping conditions and this led to different parameters entered in the gain equation and to the conclusion that differing cavity Q's accounted for the higher lasing wavelength of the flashlamp pumped case.

The following year Bass and Weber [38] again used dye laser rate and gain equations. This time they assumed a rather long Gaussian shaped pumping pulse and included triplet state effects in their discussions. They still concluded that cavity Q was the main determinant of lasing wavelength, however, they conceded that determination of lasing wavelength could be more complex if triplet state effects were significant.

In mid 1970 Keller [39] solved for pseudo steady state solutions to the rate equations (all time derivatives were set equal to zero) to investigate the effects of quenching agents on singlet and triplet state populations. He arrived at the conclusion that a specific quencher for the triplet state would markedly improve laser efficiency and that a substance that quenched both singlet and triplet states would usually improve the efficiency.

The next group to use rate equations was McColgin, et al. [33]. They considered triplet state effects but only under steady state conditions as is found in some continuous wave dye lasers, and they concluded that the emission wavelength adjusts itself so that the ratio of mirror losses, scattering losses, etc. to singlet absorption losses remains approximately constant.

Pappalardo, Samuelson and Lempicki [40] in 1972 used the rate equations to calculate the efficiency of dye lasers as a function of pump

parameters and triplet state lifetime for pump pulses in excess of one microsecond in duration. They assumed a Gaussian shaped pumping pulse and predicted that long pulse operation of up to 30 microseconds was feasible for short triplet state lifetimes but that the efficiency would go down as the pumping pulse was lengthened.

The most recent use of rate equations has been by Strome and Tuccio [17] who used results to improve their original dye laser [16], by Atkinson and Pace [34] who used them to calculate the lineshape of a Fabry-Perot etalon tuned Rhodamine 6-G dye laser, and by Streifer and Saltz [41] who used the equations to analyze an acoustooptically tuned dye laser.

B. GAIN AND RATE EQUATION PARAMETERS

The evaluation of the dye laser system rate and gain equations for a cavity such as the one in Figure 2 requires a lot of physical data. The actual dye used to obtain the experimental data had a mixture of methanol and water as the dye solvent; however, there is no information available on the dye in this particular solvent hence physical data available for ethanol solutions was used. It was assumed that any predictions or conclusions based on the results would apply to Rhodamine 6-G in the actual solvent. (The main difference in the solvents is that both the singlet absorption and fluorescence spectrum peaks are further in the green region of the visible spectrum when the solvent is 100 percent ethanol.)

The fluorescence lineshape and singlet absorption cross section of a 10^{-4} Molar solution of Rhodamine 6-G in ethanol are shown in Figure 7. These curves were obtained by F. Grum of Kodak Research Laboratories and were reproduced in Reference 8. Snavely [8] measured the fluorescence yield of the dye and obtained a value of $\phi = 0.83$ and the fluorescence spectrum of Figure 7 is normalized to this value; i.e.

$$\int_{-\infty}^{\infty} E(\lambda)\, d\lambda = \phi = 0.83.$$

There is no data available on the triplet absorption cross section for an alcohol solution of Rhodamine 6-G. The only available curve is shown in Figure 8 which was determined by Buettner [42] using a flash photolysis technique for Rhodamine 6-G in polymethyl methacrylate.

Several values of singlet and triplet state lifetimes are given in the literature. The various values given for T_s are 7.4 nsec in Weber and Bass [38], 4.8 nsec in McColgin, et al. [33], 7.3 nsec in Snavely and Peterson [31], and 5.5 nsec in Mack [43]. The reason for the use of different values by the groups mentioned above is due to different measured values. An average value of 6.3 nsec was used in this analysis because there were no significant reasons why one value should be preferred.

There are also many values of T_t given in the literature, but the best evidence available suggests that the most reasonable value is 100 nsec. Keller [39] arrived at this value in his analysis of Oxygen quenching in an alcohol solution and Snavely and Schafer [9] obtained 100 nsec as a measured value.

The value of the intersystem crossing rate constant, k_{st}, can be determined from the formula

$$k_{st}^{-1} = T_s\phi/(1 - \phi) \qquad (11)$$

which is found in Snavely [8] and others. The value of 0.84 from Bass and Steinfeld [44] is the most common value found for ϕ and since the value of 6.3 nsec is assumed for T_s, (11) yields the value of 29.4 nsec for k_{st}^{-1}. The fluorescence spectrum of Figure 7 is normalized using ϕ = 0.83 so that spectrum and equation (11) are compatible.

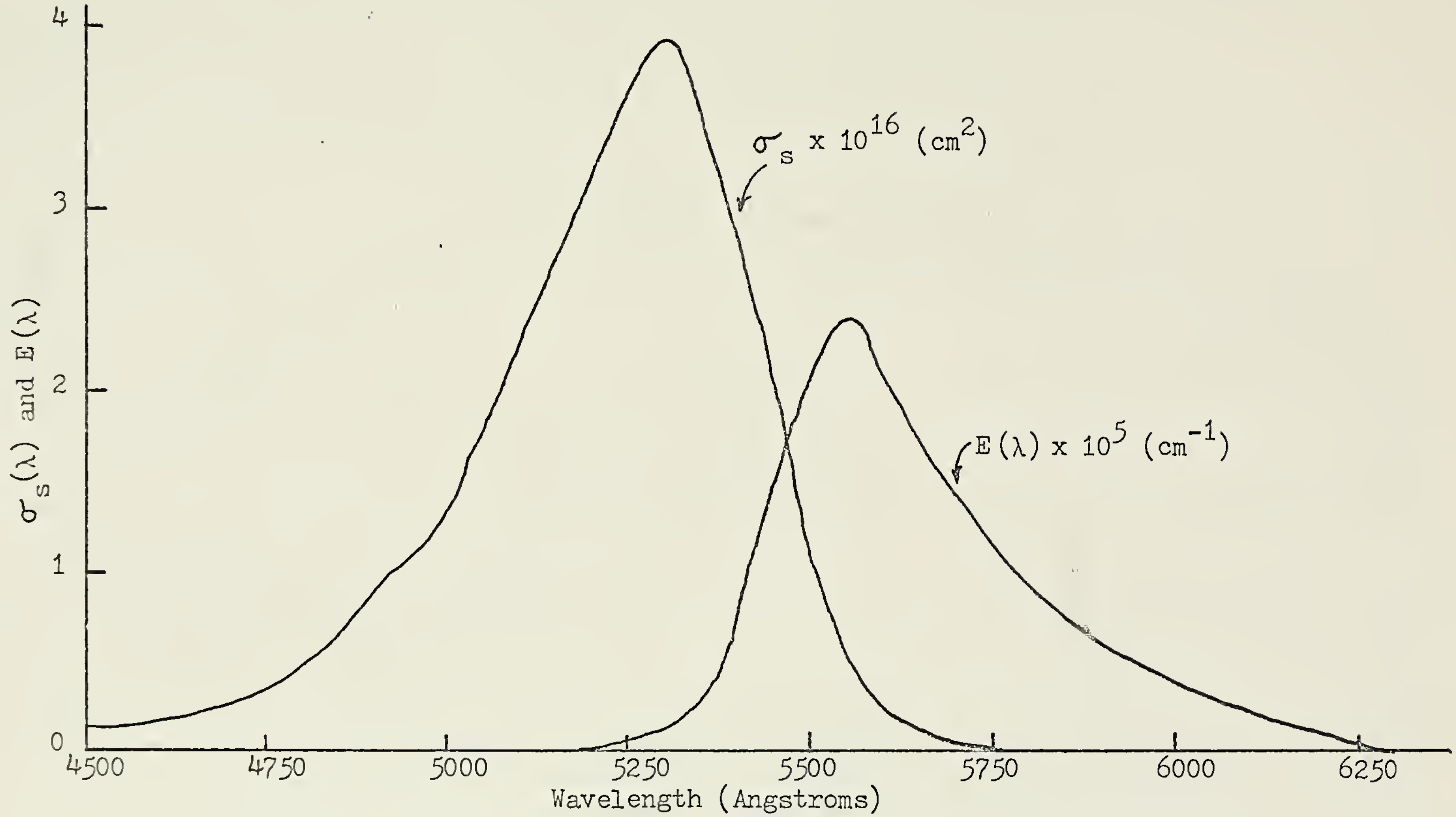


Figure 7: Singlet state absorption cross section and fluorescence spectrum of 10^{-4} Molar Rhodamine 6-G in ethanol [8].

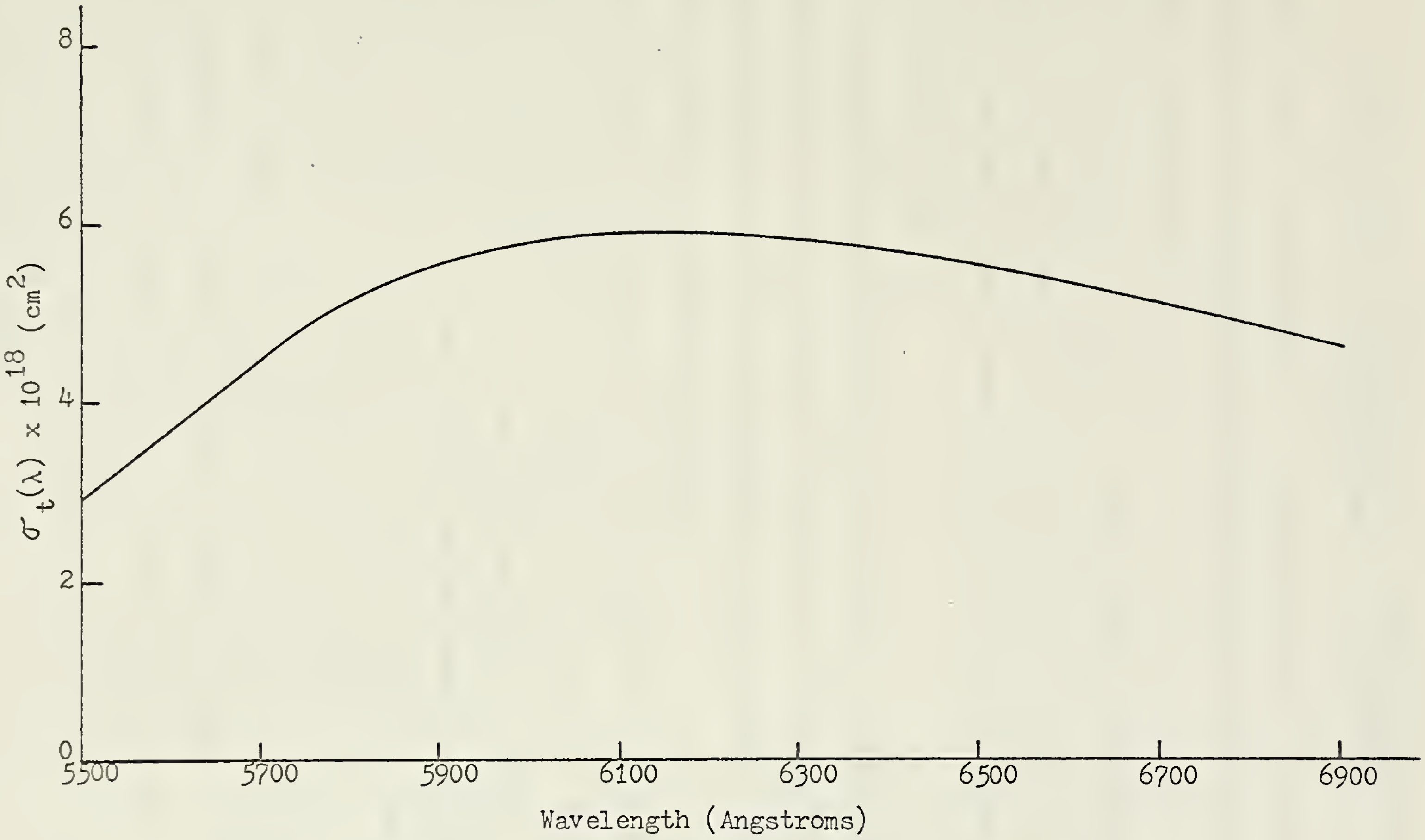


Figure 8: Triplet state absorption coefficient of Rhodamine 6-G in polymethyl methacrylate [42].

The easiest constant to evaluate is N, the total molecular density. The actual dye solution was 3×10^{-4} Molar so that

$$N = 3\text{x}10^{-4} \text{ Molar} \times \frac{1 \text{ gram-mole}}{1 \text{ Molar-liter}} \times \frac{6.023\text{x}10^{23} \text{ molecules}}{\text{gram-mole}} \times \frac{1 \text{ liter}}{10^3 \text{ cm}^3}$$

which is $1.8\text{x}10^{17}$ molecules/cm^3. This assumes that the density of the solvent at room temperature is approximately the same as at the standard reference temperature.

The values of $L_1 = .1$ cm, $R_1 = .995$ and the curve of R_2 versus wavelength were obtained from the manufacturer [45]. The dye cell windows were anti-reflection (AR) coated so typical values for AR surfaces of .98 were used for T_1 and T_2.

The remaining constants needed for evaluation of $\sigma_e(\lambda)$ are c, the speed of light, and $\bar{n}$, the index of refraction. The value used for the speed of light was $3\text{x}10^{10}$ centimeters/sec and the index of refraction of the dye solution was approximately 1.33.

The optical pumping rate, P(t), may be written as $\sigma_p I_p/hf_p$ where σ_p is the dye absorption cross section for f_p, the pump frequency, h is Plancks constant and I_p is the pump irradiance in watts/cm^2. The pump wavelength was 5145 Angstroms and the corresponding frequency was 5.82×10^{14} sec^{-1}. Plancks constant has a value of $6.625\text{x}10^{-34}$ sec^{-1}. The value of $2.37\text{x}10^{-16}$ cm^2 obtained from Figure 7 was used for σ_p.

Pump irradiance can be written as power per unit area. The model of the pump power as a function of time is shown in Figure 5. The average power output of the Argon laser was 0.5 watts and the value of 42.7 watts for P_o was obtained from the formula

$$P_{\text{average}} = \frac{1}{T}\int_o^T P(t)dt.$$

There is a problem in defining the effective area and volume in which the pump beam is absorbed. Jacobs, Samuelson, and Lempicki [46] assumed uniform pumping in their analysis of losses in a continuous wave dye laser and used the beam waist area as the effective pumped area. At least some of the Argon laser beam must exit the dye volume before uniform pumping can be assumed [40], otherwise the amount of beam penetration into the dye and the size of the pumped volume cannot be determined. The dye laser used in the experiment allowed approximately five percent of pump power to exit the dye cell [45], therefore, uniform pumping was assumed. The beam waist radius was six microns [45] so the assumed effective area was 1.13×10^{-6} cm^2.

The assumption of uniform pumping is one of the most marginal in dye laser analysis. The complexities introduced by trying to evaluate mode-matching of the pump and dye laser beams in the dye cell and nonuniform excitation of dye molecules, however, makes the assumption practical.

C. GAIN AND RATE EQUATION SOLUTIONS

1. Solution Method

The first step in the theoretical analysis was the solution of the rate equations for the state population densities as a function of time. The solution was carried out using a program entitled INTEG1 of the U.S. Naval Postgraduate School Computer Library. (See Appendix A) INTEG1 uses a fourth order Runga-Kutta method with programmable step size changes to achieve the solution to simultaneous ordinary differential equations. The only modifications made to the INTEG1 program were those necessary to enable it to handle the wide range of parameter values used in the rate equations.

The values of the state population densities were then used to evaluate the gain equation. This was accomplished by first selecting values of state densities at a particular time and then evaluating the gain using the wavelength dependent parameters over a large range of wavelengths. The succeeding steps followed the same pattern and this was continued until the point of maximum gain versus time and wavelength reached a constant relationship; i.e., the wavelength of maximum gain remained unchanged with time.

The purpose of evaluating the gain in this manner was that the lasing wavelength of the laser model should be where the gain is maximum. This was then compared to the dye fluorescence spectrum to see where the lasing wavelength was in relation to the peak wavelength of the spectrum.

The last step was to predict where the experimental laser should lase based on the theoretical comparison and the fluorescence spectrum of the experimental dye. Theoretical and experimental results should agree if theory was sound.

2. Theoretical Results

A graph of the peak gain wavelength versus time is shown in Figure 9. It can be seen from the figure that lasing starts at the longer wavelengths and sweeps toward the shorter wavelengths. Figure 9 also contains a graph of the ratio of triplet state population to ground singlet state population (N_t/N_o) versus time.

The results of evaluation of the gain versus wavelength at steady state is shown in Figure 10. It can be seen that the peak wavelength (5570 Angstroms) corresponds to the peak wavelength of the Rhodamine 6-G fluorescence curve of Figure 7.

The results displayed in Figure 9 and the gain equation, equation (9) of Section II, can be used to explain the theoretically obtained dye

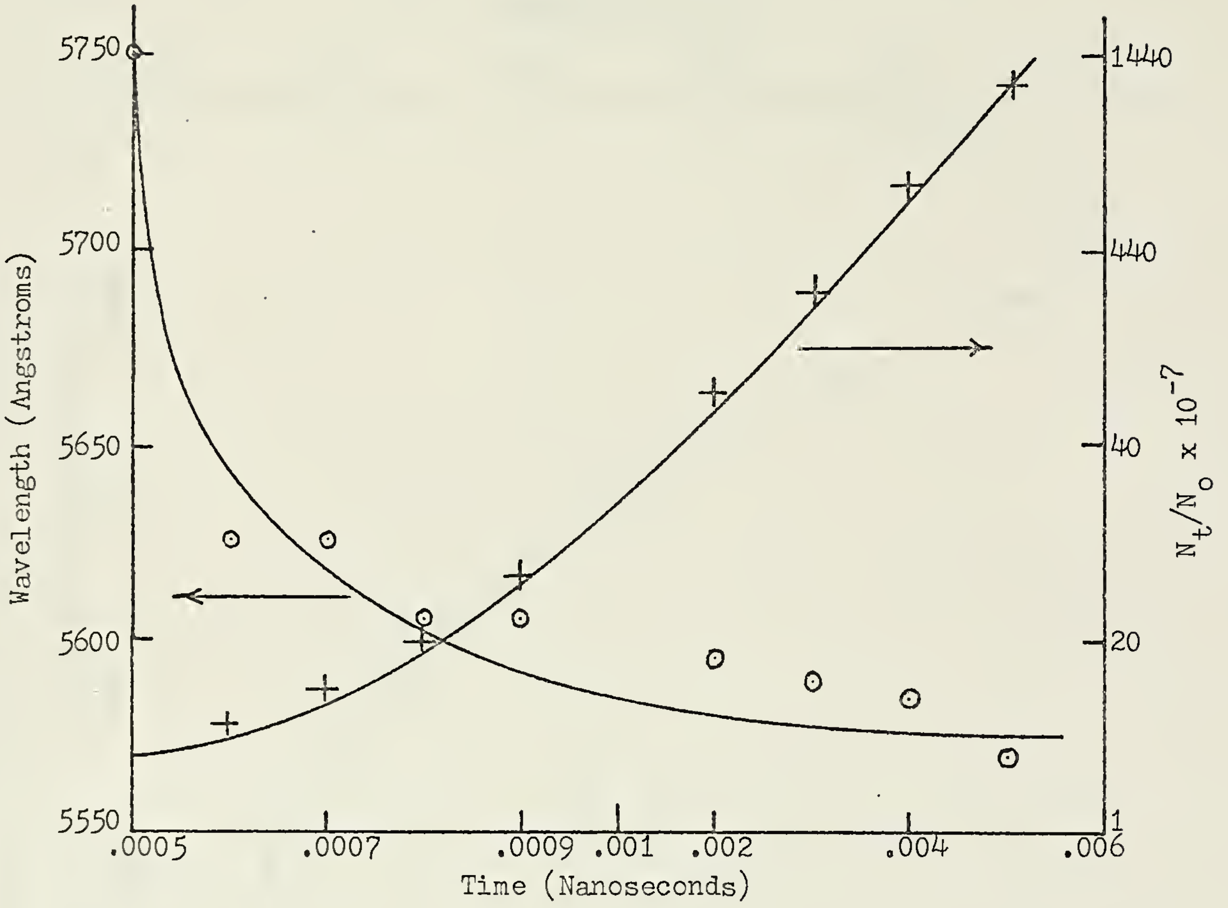


Figure 9: Peak gain wavelength and ratio of triplet to ground singlet state population for 3×10^{-4} Molar Rhodamine 6-G laser model.

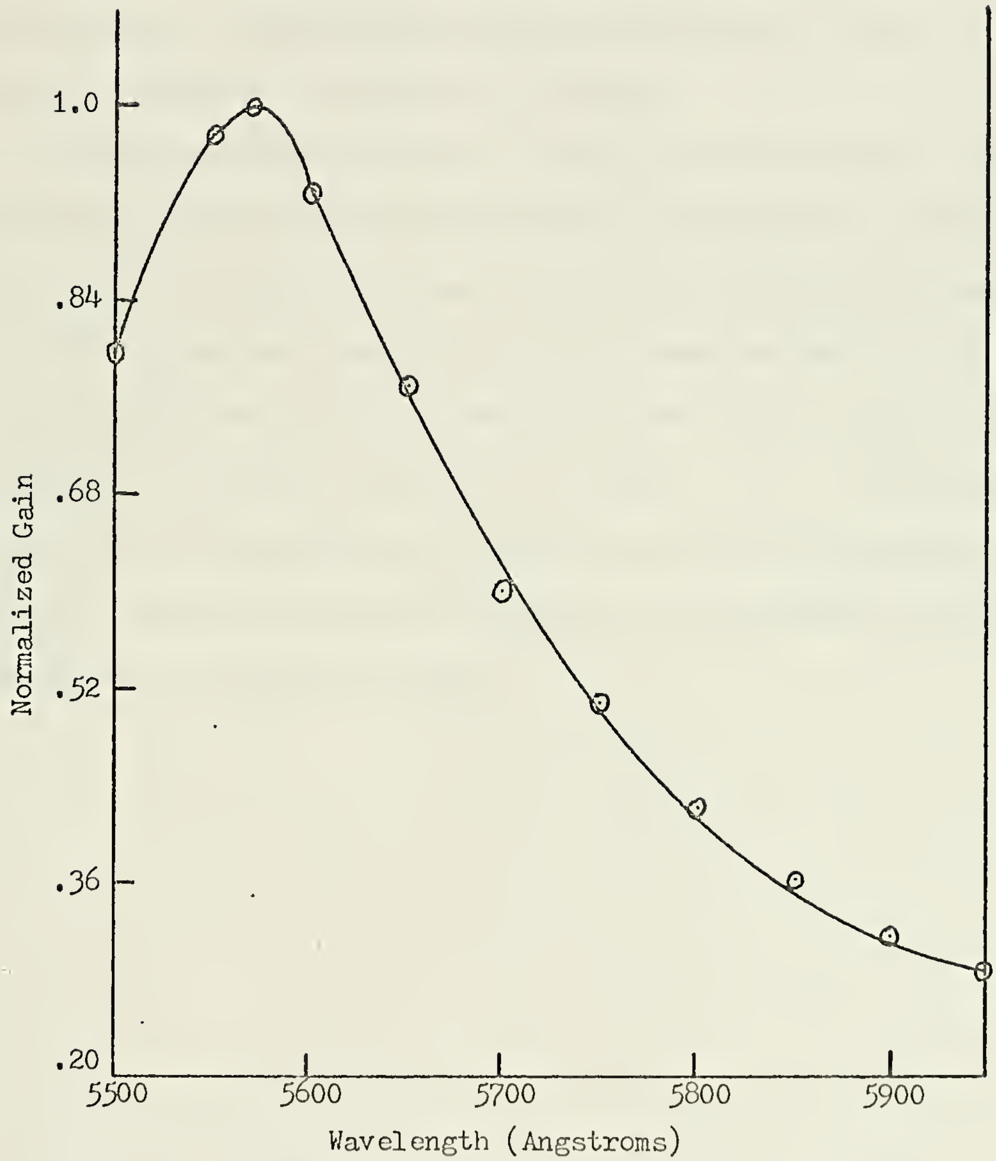


Figure 10: Theoretical normalized gain versus wavelength for Rhodamine 6-G dye laser model.

laser wavelength sweep. The two wavelength dependent losses in the gain equation are caused by singlet and triplet absorption. The ratio of N_t/N_o at the onset of lasing is very small indicating domination of the singlet state absorption loss. (Singlet absorption is associated with N_o and triplet absorption with N_t.) Figure 7 shows that singlet absorption is greatest for shorter wavelengths, hence there should be a tendency for long lasing wavelengths at the onset of lasing.

As pumping continues the ratio of N_t/N_o becomes greater. This indicates that as time passes there should be a tendency for triplet absorption losses to dominate. Figure 8 shows that triplet absorption is greater at the longer wavelengths (up to 6000 Angstroms which is within the lasing range of Rhodamine 6-G) and, therefore, the dye laser should end up lasing at wavelengths closer to the peak of the dye fluorescence curve. (The above statements are true in this case for the specific pulse model of Figure 5. Pulses of different shape, duration, and intensity may or may not cause the sweep.)

V. CONCLUSIONS

A. EXPERIMENTAL AND THEORETICAL COMPARISON

The theoretical analysis predicted that for the pumping pulse model of Figure 5 and after a wavelength sweep of very short duration the dye laser would lase at or near the peak of the dye fluorescence spectrum. It was observed experimentally that the dye laser lased approximately 50 Angstroms from the peak of the dye fluorescence curve in the pulsed case (Figure 6) rather than 200 Angstroms as in the continuous wave pumped case (Figures 4 and 6). It may be concluded, therefore, that theory and experimentation are in agreement within the limits of the assumptions made in the study.

The variable of least confidence in the entire analysis was the experimental dye fluorescence curve. The peak of the fluorescence curve for Rhodamine 6-G in an ethanol solution is typically about 5570 Angstroms [8] and in a water solution is about 5750 Angstroms [46]. The actual solvent was determined by nuclear magnetic resonance measurements to be approximately two-thirds water and one-third mathanol. This should have caused the fluorescence peak of the dye to be somewhere around 5660 to 5700 Angstroms which is much closer to the observed peak wavelength than the 5600 Angstrom peak of the fluorescence curve of Figure 6. One explanation for the difference between the measured and expected fluorescence peaks is the fact that the spectrometer calibration curve used to measure the experimental dye fluorescence spectrum had a steep slope and was very sensitive to spectrometer dial readings.

B. REMARKS

The decrease in efficiency of the dye laser as the pulse length was widened can be explained very simply in terms of the rate equations. The lengthening of the pulse decreased the peak power of the pumping pulse which in turn caused a longer time to reach the onset of lasing. As a result, the triplet state had more time to build up and extract molecules from the singlet lasing system - efficiency had to decrease. There are several things which hinder the analysis of the dye laser. There is not much data available on dye properties in various solvents and the properties of the triplet state are still pretty much unknown. There is also a lot to be learned about the effects of thermal heating and nonuniform pumping of the dye and no one as yet has begun to investigate the effect of chemical reaction and relaxation rates on the dye laser. Stability and noise considerations also require investigation.

APPENDIX A

DYE LASER RATE EQUATION COMPUTER PROGRAM LISTING

The computer listing on the following pages contains the program for the solution of the dye laser rate equations. The main part of the program is SUBROUTINE INTEG1 and the comments preceding its operational statements completely describe its use. The rate equations in the program correspond to equations (10A), (10B) and (10D) of Section II. The data cards used in the actual solution and described in the usage comments of INTEG1 are located at the end of the listing.

INTEG1 had to be modified to some extent so it could handle a wider range of parameter values than was originally allowed. These modifications included input and output format and an increase in the number of allowed integration steps and pages of output, but no changes were made to the basic fourth order Runga-Kutta integration and other computational algorithms.

DYE LASER RATE EQUATION SOLUTION

```
//SNY92083 JOB (2083,0290,DC12),'SNYDER, G. W.',TIME=2
// EXEC FORTCLG,REGION.GO=100K
//FORT.SYSIN DD *
      DIMENSION X(30),XDOT(30),C(15)
      C(10)=1.0
    1 CALL INTEG4(T,X,XDOT,C)
      IF(T.GE.0.001) GO TO 2
      C(11) = 100
      C(12) = 100
      GO TO 3
    2 C(11) = 1
      C(12) = 1
      IF(T.LE.0.1) GO TO 3
      C(11) = 100
      C(12) = 100
    3 CONTINUE
      Y = POWER(T)
C
C
C           THE THREE EQUATIONS BELOW ARE THE DYE LASER RATE EQUATIONS AND
C     CORRESPOND TO EQUATIONS 10A, 10B AND 10D OF SECTION III.  IN THE
C     EQUATIONS X(1) IS THE EXCITED SINGLET STATE POPULATION, X(2) IS THE
C     TRIPLET STATE POPULATION AND X(3) IS THE GROUND SINGLET STATE POPULATION.
C
C
      XDOT(1) = -C(2)*X(1) + C(6)*C(3)*C(7)*C(7)*Y - .542*Y*X(1) -
     1.542*Y*X(2)
      XDOT(2) = - C(4)*X(2) + C(5)*X(1)
      X(3) = 18.0*C(7)*C(7)*C(7)*C(7) - X(1) -X(2)
      GO TO 1
      END
C
C
C          FUNCTION POWER (Z) BELOW IS THE ARGON LASER POWER AS A FUNCTION OF
C     TIME IN ACCORDANCE WITH THE TWO SEGMENT CURVE OF FIGURE 5.


      FUNCTION POWER(Z)
      IF(Z.GE.12.0) GO TO 10
      POWER = (42.7*Z)/12.0
   10 POWER = 42.7*EXP((12.0-Z)/6.0)
      RETURN
      END
```

```
      SUBROUTINE INTEG4(/TC/,/XC/,/DX/,/C/)
C.......................................................................INT10010
C                                                                       INT10020
C     SUBROUTINE INTEG1 (MODIFIED VERSION CALLED INTEG4)
C                                                                       INT10040
C     PURPOSE                                                           INT10050
C                                                                       INT10060
C        INTEG1 PROVIDES A SIMPLE BUT FLEXIBLE MEANS FOR RUNGA-KUTTA    INT10070
C        SOLUTION OF ORDINARY DIFFERENTIAL EQUATIONS. INPUT AND OUTPUT  INT10080
C        ROUTINES ARE BUILT-IN ALONG WITH PRINTER POINT PLOTTING ROU-   INT10090
C        TINES. THE INTEGRATION STEP-SIZE FOR THE FOURTH ORDER NUMERI-  INT10100
C        CAL INTEGRATION CAN BE CHANGED BY APPROPRIATE DATA INPUT       INT10110
C        AND/OR PROGRAMMING. THE SUBROUTINE IS USUALLY USED FOR EQUA-   INT10120
C        TIONS WITH PREDETERMINED PARAMETERS BUT CAN BE USED TO SOLVE   INT10130
C        MORE COMPLEX PROBLEMS.                                         INT10140
C                                                                       INT10150
C     USAGE                                                             INT10160
C                                                                       INT10170
C        INTEG1 HAS BEEN COMPILED AND ADDED TO THE FORTRAN LIBRARY      INT10180
C        (MPSLIB). TO USE IT, ONE NEED ONLY SUBMIT A STANDARD OS/360    INT10190
C        PROGRAM DECK AS FOLLOWS:                                       INT10200
C                                                                       INT10210
C        //  ---- STANDARD JOB CARD ----                                INT10220
C        //  EXEC  FORTCLG,REGION.GO=100K                               INT10230
C        //FORT.SYSIN DD *                                              INT10240
C              DIMENSION X(30),XDOT(30),C(15)                           INT10250
C              C(10)=1.                                                 INT10260
C            1 CALL INTEG1(T,X,XDOT,C)                                  INT10270
C                                                                       INT10280
C              *  *  *  *  *  *  *  *  *  *  *  *                       INT10290
C              *                             *                          INT10300
C              * FORTRAN EQUATIONS TO BE SOLVED *                       INT10310
C              *                             *                          INT10320
C              *  *  *  *  *  *  *  *  *  *  *  *                       INT10330
C                                                                       INT10340
C              GO TO 1                                                  INT10350
C              END                                                      INT10360
C        /*                                                             INT10370
C        //GO.SYSIN DD *                                                INT10380
C                                                                       INT10390
C              *  *  *  *  *  *  *  *  *  *  *  *                       INT10400
C              *                             *                          INT10410
C              * DATA CARDS AS DESCRIBED BELOW *                        INT10420
C              *                             *                          INT10430
C              *  *  *  *  *  *  *  *  *  *  *  *                       INT10440
C                                                                       INT10450
C        /*  (ORANGE)                                                   INT10460
C                                                                       INT10470
```

```
C     EQUATION STATEMENTS                                               INT10480
C                                                                       INT10490
C         THE NUMBER OF EQUATIONS TO BE SOLVED, "N", MUST BE LESS THAN  INT10500
C         OR EQUAL 30. THEY MUST BE REDUCED TO STANDARD FIRST ORDER     INT10510
C         FORM AND THEN EXPRESSED IN FORTRAN. ADDITIONAL NONSUBSCRIPTED INT10520
C         VARIABLES MAY BE USED AS WELL AS STANDARD LIBRARY FUNCTIONS   INT10530
C         AND/OR SUBROUTINES. THE ORDER OF THE EQUATIONS IS USUALLY     INT10540
C         UNIMPORTANT. A TYPICAL SET MIGHT READ:                        INT10550
C                                                                       INT10560
C               ERROR=SIN(C(1)*T)-X(1)                                  INT10570
C               XDOT(1)=X(2)-.1*X(1)                                    INT10580
C               XDOT(2)=ERROR+C(2)*XDOT(1)                              INT10590
C                                                                       INT10600
C         THE X ARRAY CONTAINS THE DEPENDENT VARIABLES.  THE XDOT ARRAY INT10610
C         CONTAINS THE FIRST DERIVATIVE WITH RESPECT TO THE INDEPENDENT INT10620
C         VARIABLE ("T" IN THIS EXAMPLE). THE EQUATIONS TO BE SUPPLIED  INT10630
C         BY THE USER DEFINE EACH "XDOT" IN TERMS OF THE INDEPENDENT    INT10640
C         AND DEPENDENT VARIABLES, CONSTANTS AND OTHER VALUES IN THE    INT10650
C         "XDOT" ARRAY.                                                 INT10660
C                                                                       INT10670
C         CONSTANTS (C(I),I.LE.8) ARE READ FROM A DATA CARD. CONSTANT   INT10680
C         C(10) MUST NEVER BE USED EXCEPT AS IN THE SAMPLE DECK SETUP   INT10690
C         ABOVE. CONSTANTS C(11),C(12), AND C(13) CONTROL FREQUENCY OF  INT10700
C         PRINT-OUT DURING INTEGRATION, THE FREQUENCY OF PLOT POINT     INT10710
C         GENERATION, AND THE INTEGRATION STEP-SIZE (IN THAT ORDER).    INT10720
C         THE DEFAULTS ARE PRINT-OUT EVERY 20TH STEP AND PLOT POINTS    INT10730
C         GENERATED AT EACH 5TH STEP. THE INTEGRATION STEP-SIZE IS      INT10740
C         DEFINED ON A DATA CARD. THESE CONSTANTS MAY BE CHANGED BY     INT10750
C         REDEFINING THEM BETWEEN THE "CALL INTEG1" AND "GO TO 1" STATE-INT10760
C         MENTS ABOVE (AT ANY POINT DURING THE SOLUTION) SUCH AS:       INT10770
C                                                                       INT10780
C               C(11)=10.                                               INT10790
C               C(12)=1.                                                INT10800
C                                                                       INT10810
C         CAUSES PRINT-OUT TO TAKE PLACE AT EVERY 10 STEPS AND PLOT     INT10820
C         POINTS TO BE GENERATED AT EACH STEP. NOTE THAT NO MORE THAN   INT10830
C         4500 INTEGRATION STEPS, 450 LINES OF PRINT-OUT, OR 900 PLOT   INT10840
C         POINTS CAN BE GENERATED IN ANY ONE RUN.                       INT10850
C                                                                       INT10860
C         ANY LEGAL FORTRAN STATEMENT OR TECHNIQUE CAN BE USED IN THE   INT10870
C         CALLING PROGRAM, PROVIDED THAT A TRANSFER OF CONTROL DOES NOT INT10880
C         PREVENT THE PROCESSING OF ALL RELEVANT STATEMENTS DURING EACH INT10890
C         STEP OF THE INTEGRATION. FOR EXAMPLE THE FOLLOWING WOULD BE   INT10900
C         ALLOWED:                                                      INT10910
C                                                                       INT10920
C               IF(T-10.0) 3,2,2                                        INT10930
C             3 C(1)=0.                                                 INT10940
C             2 CONTINUE                                                INT10950
```

```
C                                                                       INT10960
C          WHICH CAUSES C(1) TO TAKE THE VALUE SPECIFIED ON THE DATA CARDINT10970
C          FOR T.LT.10. AND THE VALUE 0. FOR T.GE.10. ONLY QUANTITIES   INT10980
C          X(I) AND T CAN BE OUTPUT. IF SOME QUANTITY OTHER THAN THE    INT10990
C          INDEPENDENT VARIABLE, DEPENDENT VARIABLE OR DERIVATIVE       INT11000
C          IS DESIRED AS OUTPUT, IT MUST BE EQUATED TO AN X(I) SUCH THAT INT11010
C          N.LT.I.LE.30. SUCH AS:                                       INT11020
C                                                                       INT11030
C               X(3)=ERROR                                              INT11040
C               X(4)=ERROR*ERROR.                                       INT11050
C                                                                       INT11060
C          ALL SUCH AUXILLIARY EQUATIONS MUST BE PLACED BETWEEN THE     INT11070
C          "CALL INTEG1" AND "GO TO 1" STATEMENTS.                      INT11080
C                                                                       INT11090
C       DATA CARDS                                                      INT11100
C                                                                       INT11110
C       FIRST:    CONTAINS A JOB IDENTIFICATION LABEL PUNCHED IN COLS 1 TOINT11120
C                 48. IT WILL BE REPRODUCED IN THE PRINT-OUT.           INT11130
C                                                                       INT11140
C       SECOND:   CONTAINS THE NUMBER OF RUNS TO BE PROCESSED (.LE.9)   INT11150
C                 PUNCHED IN COL 1. THE RUN NUMBER IS PLACED ON ALL OUTPUTINT11160
C                 ALONG WITH THE ABOVE JOB IDENTIFICATION.              INT11170
C                                                                       INT11180
C       THIRD:    CONTAINS THE VALUE OF N ,THE NUMBER OF EQUATIONS TO BE INT11190
C                 SOLVED. PUNCHED RIGHT-JUSTIFIED IN COLS 1 AND 2.      INT11200
C                 WHEN N IS LESS THAN 10, IT MUST BE PUNCHED IN COL 2.  INT11210
C                 WHEN IT IS EQUAL TO OR GREATER THAN 10 IT MUST BE PUNCH-INT11220
C                 ED IN COLS 1 AND 2. IT MUST BE LESS THAN OR EQUAL TO 30.INT11230
C                                                                       INT11240
C       FOURTH:   INTRODUCES THE INITIAL AND FINAL TIMES OF T (TI & TF) INT11250
C                 TOGETHER WITH THE STEP-SIZE(S), DT. THE INTEGRATION CAN INT11260
C                 BE PROCESSED IN UP TO 3 SEGMENTS, EACH WITH A DIFFERENT INT11270
C                 STEP-SIZE, THUS THE DATA CARD CAN CONTAIN:            INT11280
C                                                                       INT11290
C                     TI - DT1 - TF                                     INT11300
C                     TI - DT1 - TF1 - DT2 - TF                         INT11310
C                  OR TI - DT1 - TF1 - DT2 - TF2 - DT3 - TF             INT11320
C                                                                       INT11330
C                 THE CORRESPONDING DATA VALUES ARE PUNCHED (WITH DECI- INT11340
C                 MALS), IN THE ABOVE ORDER, IN COLS 1-10,11-20,21-30,  INT11350
C                 ...,61-70.                                            INT11360
C                                                                       INT11370
C       FIFTH:    CONTAINS THE CONSTANTS C(1) THRU C(8) PUNCHED (WITH   INT11380
C                 DECIMALS) IN COLS 1-10,11-20,...,71-80. BLANK MAY BE  INT11390
C                 USED FOR EITHER ZERO OR UNUSED CONSTANTS.             INT11400
C                                                                       INT11410
C       SIXTH:    CONTAINS THE INITIAL CONDITIONS (X(1)(0)) THROUGH     INT11420
C                 (X(N)(0)) IN THE SAME FORMAT AS THE FIFTH CARD. ADDI- INT11430
```

```
C              TIONAL CARDS ARE REQUIRED IF N.GT.8.                     INT11440
C                                                                       INT11450
C    NEXT TO                                                            INT11460
C    LAST:     CONTROLS THE PRINT-OUT. A BLANK CARD WILL SUPPRESS THE   INT11470
C              OUTPUT. UP TO 8 QUANTITIES MAY BE PRINTED OUT DURING     INT11480
C              INTEGRATION. EACH QUANTITY IS IDENTIFIED BY A 1 TO 8     INT11490
C              CHARACTER COLUMN HEADING AND A 2-DIGIT SUBSCRIPT OF      INT11500
C              THE VARIABLE TO BE PRINTED UNDER THE HEADING. THE FIRST  INT11510
C              COLUMN HEADING IS PUNCHED IN COLS 1-8 AND THE CORRES-    INT11520
C              PONDING SUBSCRIPT IN COLS 9-10. THE SECOND COLUMN HEAD-  INT11530
C              ING IS PUNCHED IN COLS 11-18 AND THE SUBSCRIPT IN 19-20.INT11540
C              CONTINUE IN THIS MANNER UP TO A MAXIMUM OF 8 QUANTITIES.INT11550
C              THE SUBSCRIPT ZERO (OR BLANK) IS USED TO SPECIFY THE     INT11560
C              INDEPENDENT VARIABLE, T.                                 INT11570
C                                                                       INT11580
C    LAST:     CONTROLS THE PLOT OUTPUT. A BLANK CARD WILL SUPPRESS     INT11590
C              THE OUTPUT. UP TO 4 CURVES CAN BE PLOTTED. THEY CAN ALL  INT11600
C              BE PLOTTED ON ONE GRAPH OR ALL ON SEPARATE GRAPHS. EACH  INT11610
C              CURVE IS SPECIFIED BY A POINT-PLOT TITLE OF 1-16 CHAR-   INT11620
C              ACTERS, THE SUBSCRIPT OF THE Y-ORDINATE (2 DIGITS), AND  INT11630
C              THE SUBSCRIPT OF THE X-ORDINATE (2 DIGITS). THE FIRST    INT11640
C              TITLE IS PUNCHED IN COLS 1-16, THE FIRST Y-ORDINATE IN   INT11650
C              COLS 17-18, AND THE FIRST X-ORDINATE IN COLS 19-20. THE  INT11660
C              SECOND TITLE IS IN COLS 21-36, THE SECOND Y-ORDINATE IN  INT11670
C              COLS 37-38, AND THE SECOND Y-ORDINATE IN COLS 39-40,     INT11680
C              AND SO ON UP THRU 4 GRAPHS. WHEN ALL CURVES ARE TO BE    INT11690
C              ON ONE GRAPH, SPECIFY ONLY THE FIRST TITLE AND LEAVE     INT11700
C              THE OTHER TITLE FIELDS BLANK.                            INT11710
C                                                                       INT11720
C              INTEG1 USES SUBROUTINES PLOTP AND UTPLOT TO GENERATE     INT11730
C              THE POINT PLOTTING.                                      INT11740
C                                                                       INT11750
C    MULTIPLE RUNS                                                      INT11760
C                                                                       INT11770
C        IF SEVERAL SOLUTIONS OF THE SAME EQUATIONS ARE DESIRED, THE    INT11780
C        NUMBER OF RUNS IS SPECIFIED ON THE SECOND DATA CARD. FOR EACH  INT11790
C        RUN AFTER THE FIRST, ONLY THE FOURTH THRU THE LAST CARDS ARE   INT11800
C        REQUIRED. NO INFORMATION, BESIDES THE FIRST 3 CARDS, IS        INT11810
C        RETAINED BETWEEN RUNS.                                         INT11820
C                                                                       INT11830
C    CAUTIONS TO USER                                                   INT11840
C                                                                       INT11850
C        TOO LARGE A STEP-SIZE CAN RESULT IN UNACCEPTABLE INTEGRATION   INT11860
C        ERRORS AND EVEN INSTABILITY OF THE INTEGRATION PROCESS. THE    INT11870
C        ONLY (MODERATELY) SAFE TEST IS TO RUN A PROBLEM WITH AT LEAST  INT11880
C        TWO DIFFERENT STEP-SIZES AND THEN COMPARE THE RESULTS.         INT11890
C        VALUES OF STEP-SIZE LARGER THAN UNITY ARE USUALLY NOT          INT11900
C        RECOMMENDED.                                                   INT11910
```

```
C                                                                       INT11920
C     PROGRAMMER                                                        INT11930
C         J. R. WARD, OCTOBER 1963                                      INT11940
C         REVISED 6/64,6/65,6/67,11/71                                  INT11950
C                                                                       INT11960
C.......................................................................INT11970
C                                                                       INT11980
      REAL*8 ITITLE(12),JTITLE(8),IBLANK/'        '/,
     1KTITLE(8)
      DIMENSION X(30),BX(30),XC(30),C(15),IP(10),IG(10),FR(10),GR(10),  INT12010
     1TX(5),TY(5),X1(900),Y1(900),X2(900),Y2(900),X3(900),Y3(900),      INT12020
     2X4(900),Y4(900)                                                   INT12030
      REAL LABEL,RUN(2)/' RUN','    '/,BT(9)/' 1  ',' 2  ',' 3  ',' 4  'INT12040
     1,' 5  ',' 6  ',' 7  ',' 8  ',' 9  '/                              INT12050
      EQUIVALENCE (ITITLE(7),RUN(1))                                    INT12060
      INDIC = C(10)+0.0000001                                           INT12070
      GO TO (1, 2000, 50, 58, 88, 88),INDIC                             INT12080
C                                                                       INT12090
C         READ DATA AND PRINT RECORD.                                   INT12100
C                                                                       INT12110
    1 READ(5,100) (ITITLE(I), I=1,6)                                    INT12120
  100 FORMAT (10A8)                                                     INT12130
      READ (5,101)NR                                                    INT12140
  101 FORMAT (I1)                                                       INT12150
      READ (5,102)NN                                                    INT12160
  102 FORMAT (I2)                                                       INT12170
      NRC = 0                                                           INT12180
      IF(NN.LE.30) GO TO 1000                                           INT12190
      WRITE(6,200)                                                      INT12200
  200 FORMAT (////,48H ERROR IN ORDER OF EQUATION. MUST NOT EXCEED 30. )INT12210
      STOP                                                              INT12220
 1000 NRC = NRC + 1                                                     INT12230
      WRITE (6,201) (ITITLE(I),I=1,6)                                   INT12240
  201 FORMAT (1H1,/////,36X,6A8)                                        INT12250
      IF(NRC.EQ.1.AND.NR.EQ.1) GO TO 5                                  INT12260
      WRITE(6,202)NR                                                    INT12270
  202 FORMAT (/,37X,I1,20H RUNS ARE CALLED FOR )                        INT12280
      GO TO 6                                                           INT12290
    5 WRITE(6,203)                                                      INT12300
  203 FORMAT (/,37X,21HONE RUN IS CALLED FOR ,///,18H INPUT DATA RECORD)INT12310
      GO TO 7                                                           INT12320
    6 WRITE(6,204)NRC                                                   INT12330
  204 FORMAT (///,34H INPUT DATA RECORD FOR RUN NUMBER  ,I1)            INT12340
    7 WRITE(6,205)NN                                                    INT12350
  205 FORMAT (//,22H ORDER OF EQUATIONS = ,I2,/)                        INT12360
      READ (5,103)TI,DT,TF1,DT2,TF2
  103 FORMAT(5F15.6)
      READ (5,5000) DT3,TF3
```

```
 5000 FORMAT(2F15.6)
      TF = TF1                                                          INT12390
      IF(DT2.NE.0.) GO TO 9                                             INT12400
      WRITE(6,206)TI,TF                                                 INT12410
  206 FORMAT (22H INITIAL TIME          = ,E19.11,/
     1        22H FINAL TIME            = ,E19.11)
      WRITE(6,207)DT                                                    INT12440
  207 FORMAT (22H STEP SIZE             = ,E19.11)
      GO TO 12                                                          INT12460
    9 IF(DT3.NE.0.) GO TO 11                                            INT12470
      TF = TF2                                                          INT12480
      WRITE(6,206) TI,TF                                                INT12490
      WRITE(6,208)DT,TI,TF1,DT2,TF1,TF                                  INT12500
  208 FORMAT (22H STEP SIZE             = ,E19.11,13H BETWEEN T = ,E19.11,
     1         9H AND T = ,E19.11)
      GO TO 12                                                          INT12530
   11 TF=TF3                                                            INT12540
      WRITE(6,208) DT,TI,TF1,DT2,TF1,TF2,DT3,TF2,TF                     INT12550
   12 READ(5,5001) (C(I), I=1,8)
 5001 FORMAT (8F10.4)
      READ (5,106)(X(I),I=1,NN)
  106 FORMAT (3F25.5)
      J = 0                                                             INT12580
      DO 14 I=1,8                                                       INT12590
      IF(C(I).NE.0.) J=J+1                                              INT12600
   14 CONTINUE                                                          INT12610
      K = 0                                                             INT12620
      DO 16 I=1,NN                                                      INT12630
      IF(X(I).NE.0.) K=K+1                                              INT12640
   16 CONTINUE                                                          INT12650
      IF(J - 1)17,18,19                                                 INT12660
   17 WRITE (6,209)                                                     INT12670
  209 FORMAT (/,34H ALL THE CONSTANTS, C(I), ARE ZERO )                 INT12680
      GO TO 423                                                         INT12690
   18 WRITE (6,210)                                                     INT12700
  210 FORMAT (/,30H THE ONLY NON-ZERO CONSTANT IS )                     INT12710
      GO TO 420                                                         INT12720
   19 WRITE (6,211)                                                     INT12730
  211 FORMAT (/,35H THE NON-ZERO CONSTANTS, C(I), ARE )                 INT12740
  420 DO 422 I=1,8                                                      INT12750
      IF(C(I).NE.0.) WRITE(6,212) I,C(I)                                INT12760
  212 FORMAT (14X,2HC(,I2,4H) = ,E12.4)
  422 CONTINUE                                                          INT12780
  423 IF(K - 1)424,425,426                                              INT12790
  424 WRITE (6,1209)                                                    INT12800
 1209 FORMAT (/,36H ALL THE INITIAL CONDITIONS ARE ZERO )               INT12810
      GO TO 20                                                          INT12820
  425 WRITE (6,1210)                                                    INT12830
```

```
1210 FORMAT (/,39H THE ONLY NON-ZERO INITIAL CONDITION IS )        INT12840
     GO TO 427                                                     INT12850
 426 WRITE (6,1211)                                                INT12860
1211 FORMAT (/,36H THE NON-ZERO INITIAL CONDITIONS ARE )           INT12870
 427 DO 429 I=1,NN                                                 INT12880
     IF(X(I).NE.0.) WRITE(6,1212) I,X(I)                           INT12890
1212 FORMAT (14X,2HX(,I2,4H) = ,E25.5)                             INT12900
 429 CONTINUE                                                      INT12910
  20 READ (5,104) (JTITLE(I),IP(I),I=1,8)
 104 FORMAT(8(A8,I2))
C                                                                  INT12940
C    CHECK FOR THE NUMBER OF COLUMNS CALLED FOR BY LOCATING FIRST  INT12950
C    BLANK COLUMN HEADING                                          INT12960
C                                                                  INT12970
     DO 21 J=1,8                                                   INT12980
     IF(JTITLE(J).EQ.IBLANK) GO TO 22                              INT12990
  21 CONTINUE                                                      INT13000
     J = 9                                                         INT13010
  22 JJ = J - 1                                                    INT13020
C                                                                  INT13030
C    JJ IS NOW THE NUMBER OF COLUMNS. REPEAT WITH THE GRAPHS.       INT13040
C                                                                  INT13050
     READ (5,105)(KTITLE(I),KTITLE(I+1),IG(I),IG(I+1),I=1,7,2)     INT13060
 105 FORMAT (4(2A8,2I2))                                           INT13070
     DO 24 K=1,7,2                                                 INT13080
     IF(KTITLE(K).EQ.IBLANK.AND.KTITLE(K+1).EQ.IBLANK) GO TO 25    INT13090
  24 CONTINUE                                                      INT13100
     K = 8                                                         INT13110
  25 KK = K/2                                                      INT13120
     KKK = KK*2                                                    INT13130
     MULTIP = 0                                                    INT13140
     IF(KK.NE.1) GO TO 306                                         INT13150
     IF(IG(3) + IG(4).EQ.0) GO TO 306                              INT13160
     IF(IG(5) + IG(6).NE.0) GO TO 303                              INT13170
     MULTIP = 2                                                    INT13180
     KKK = 4                                                       INT13190
     GO TO 306                                                     INT13200
 303 IF(IG(7) + IG(8).NE.0) GO TO 305                              INT13210
     MULTIP = 3                                                    INT13220
     KKK = 6                                                       INT13230
     GO TO 306                                                     INT13240
 305 MULTIP = 4                                                    INT13250
     KKK = 8                                                       INT13260
C                                                                  INT13270
C    IF MULTIP = 0, KK IS THE NUMBER OF SINGLE CURVE GRAPHS. OTHERWISE INT13280
C    MULTIP IS THE NUMBER OF CURVES ON A SINGLE GRAPH.             INT13290
C                                                                  INT13300
 306 IF(JJ.EQ.0) GO TO 27                                          INT13310
```

```
      WRITE(6,214) (JTITLE(I),IP(I),I=1,JJ)                             INT13320
  214 FORMAT (///,56H THE COLUMN HEADINGS AND THE CORRESPONDING VARIABLEINT13330
     1S ARE ,//,(15X,A8,9X,2HX(,I2,1H)))
      GO TO 28                                                          INT13350
   27 WRITE(6,215)                                                      INT13360
  215 FORMAT (///,25H NO PRINTOUT IS REQUIRED   )                       INT13370
   28 IF(KK.EQ.0) GO TO 308                                             INT13380
      IF(MULTIP.NE.0) GO TO 309                                         INT13390
      IF(KK.NE.1) GO TO 307                                             INT13400
      WRITE (6,216) KTITLE(1),KTITLE(2),IG(1),IG(2)                     INT13410
  216 FORMAT (///,52H THE GRAPH TITLE AND THE CORRESPONDING VARIABLES ARINT13420
     1E ,//,10X,2A8,4X,2HX(,I2,8H) VS. X(,I2,1H))                       INT13430
      GO TO 31                                                          INT13440
  307 WRITE (6,217) (KTITLE(I),KTITLE(I+1),IG(I),IG(I+1),I=1,KKK,2)     INT13450
  217 FORMAT (///,64H THE INDIVIDUAL GRAPH TITLES AND THE CORRESPONDING INT13460
     1VARIABLES ARE ,//,(10X,2A8,4X,2HX(,I2,8H) VS. X(,I2,1H)))         INT13470
      GO TO 31                                                          INT13480
  308 WRITE (6,1217)                                                    INT13490
 1217 FORMAT(///,24H NO GRAPHS ARE REQUIRED   )                         INT13500
      GO TO 31                                                          INT13510
  309 WRITE(6,1220)                                                     INT13520
 1220 FORMAT (///,52H THE GRAPH TITLE AND THE CORRESPONDING VARIABLES ARINT13530
     1E ,/)                                                             INT13540
      WRITE(6,1221) KTITLE(1),KTITLE(2),(IG(I),IG(I+1),I=1,KKK,2)       INT13550
 1221 FORMAT (10X,2A8,4X,2HX(,I2,8H) VS. X(,I2,1H),/, (30X,2HX(,I2,     INT13560
     1          8H) VS. X(,I2,1H)))                                     INT13570
C                                                                       INT13580
C     THIS ENDS THE BOOK-KEEPING. INITIALIZE BEFORE ENTERING MAIN LOOP. INT13590
C                                                                       INT13600
   31 IPAGE = 0                                                         INT13610
      T = TI                                                            INT13620
      NCPTS = 0                                                         INT13630
      NUMPTS = 0                                                        INT13640
      ITITLE(9)=IBLANK                                                  INT13650
      ITITLE(11)=IBLANK                                                 INT13660
      ITITLE(12)=IBLANK                                                 INT13670
      RUN(2)=BT(NRC)                                                    INT13680
      C(11) = 20.                                                       INT13690
      C(12) = 5.                                                        INT13700
      C(13) = DT                                                        INT13710
      DO 42 I=1,NN                                                      INT13720
   42 XC(I) = X(I)                                                      INT13730
      TC = T                                                            INT13740
      C(10) = 2.                                                        INT13750
      RETURN                                                            INT13760
C                                                                       INT13770
 2000 IF(JJ.EQ.0) GO TO 54                                              INT13780
C                                                                       INT13790
```

```
      INCPR = C(11)+0.0000001                                            INT13800
      C(11) = 20.                                                        INT13810
      IF( MOD (NCPTS,50*INCPR).EQ.0) GO TO 46                            INT13820
      IF( MOD (NCPTS,10*INCPR).EQ.0) GO TO 47                            INT13830
      IF( MOD (NCPTS,    INCPR))54,48,54                                 INT13840
   46 IPAGE = IPAGE + 1                                                  INT13850
      IF(NR.EQ.1) GO TO 1047                                             INT13860
      WRITE (6,218) (ITITLE(I),I=1,6),IPAGE,ITITLE(7),(JTITLE(I),I=1,8)  INT13870
      WRITE(6,219)                                                       INT13880
      GO TO 47                                                           INT13890
 1047 WRITE (6,1218)(ITITLE(I),I=1,6),IPAGE,(JTITLE(I),I=1,8)            INT13900
      WRITE (6,219)                                                      INT13910
   47 WRITE(6,219)                                                       INT13920
  218 FORMAT (1H1,///,23X,6A8,7X,5HPAGE ,I2,14H OF OUTPUT FOR,A8,/////,
     1       21X,4(A8,17X))
 1218 FORMAT(1H1,///,23X,6A8,27X,5HPAGE ,I2,/////,21X,4(A8,17X))
  219 FORMAT (1H   )                                                     INT13960
   48 DO 49 I=1,NN                                                       INT13970
   49 XC(I) = X(I)                                                       INT13980
      TC = T                                                             INT13990
      C(10) = 3.                                                         INT14000
      RETURN                                                             INT14010
C                                                                        INT14020
   50 DO 53 I=1,JJ                                                       INT14030
C                                                                        INT14040
      PR(I) = T                                                          INT14050
      IF(IP(I).NE.0) PR(I)=XC(IP(I))                                     INT14060
   53 CONTINUE                                                           INT14070
      WRITE (6,220)(PR(I),I=1,JJ)                                        INT14080
  220 FORMAT (7X, 8E25.5)                                                INT14090
   54 IF(KK.EQ.0) GO TO 62                                               INT14100
      INCGR = C(12)+0.0000001                                            INT14110
      C(12) = 5.                                                         INT14120
      IF( MOD (NCPTS, INCGR).NE.0) GO TO 62                              INT14130
      DO 57 I=1,NN                                                       INT14140
   57 XC(I) = X(I)                                                       INT14150
      TC = T                                                             INT14160
      C(10) = 4.                                                         INT14170
      RETURN                                                             INT14180
C                                                                        INT14190
   58 DO 61 I=1,KKK                                                      INT14200
C                                                                        INT14210
      GR(I) = T                                                          INT14220
      IF(IG(I).NE.0) GR(I)=XC(IG(I))                                     INT14230
   61 CONTINUE                                                           INT14240
      IF(KKK.GE.8) GO TO 1610                                            INT14250
      KP1 = KKK + 1                                                      INT14260
      DO 1612 I=KP1,8                                                    INT14270
```

```
1612 GR(I) = 0.                                                              INT14280
1610 NUMPTS = NUMPTS + 1                                                      INT14290
     Y1(NUMPTS) = GR(1)                                                       INT14300
     X1(NUMPTS) = GR(2)                                                       INT14310
     Y2(NUMPTS) = GR(3)                                                       INT14320
     X2(NUMPTS) = GR(4)                                                       INT14330
     Y3(NUMPTS) = GR(5)                                                       INT14340
     X3(NUMPTS) = GR(6)                                                       INT14350
     Y4(NUMPTS) = GR(7)                                                       INT14360
     X4(NUMPTS) = GR(8)                                                       INT14370
  62 NOPTS = NOPTS + 1                                                        INT14380
     IF(NUMPTS.LT.1000) GO TO 64
     WRITE (6,221)                                                            INT14400
 221 FORMAT (//////,26H STOP AT 1000 GRAPH POINTS     )
     GO TO 91                                                                 INT14420
  64 IF(NOPTS.LT.10000) GO TO 66
     WRITE (6,222)                                                            INT14440
 222 FORMAT (//////,32H STOP AT 10000 INTEGRATION STEPS )                     IN
     GO TO 91                                                                 INT14460
  66 IF(IPAGE - 15) 69,67,68
  67 IF( MOD (NOPTS , 50*INCPR).NE.0) GO TO 69                                INT14480
  68 WRITE (6,223)                                                            INT14490
 223 FORMAT(//////,27H STOP AT 10 PAGES OF OUTPUT    )
     GO TO 91                                                                 INT14510
  69 DO 70 I=1,NN                                                             INT14520
     IF(ABS(X(I)).GT.1.0E+18) GO TO 71
  70 CONTINUE                                                                 INT14540
     GO TO 72                                                                 INT14550
  71 WRITE (6,224)                                                            INT14560
 224 FORMAT (//////,76H STOP WITH THE ABSOLUTE VALUE OF A DEPENDENT VARINT14570
    1IABLE GREATER THAN 1.0E+18.  ,/,57H INTEGRATION PROBABLY UNSTABLE.INT44580
    2 TRY A SMALLER STEP SIZE. ,26HNO GRAPHS WILL BE PLOTTED.        )  INT14590
     GO TO 330                                                                INT14600
  72 DT = C(13)                                                               INT14610
     IF(TI.GT.TF) GO TO 80                                                    INT14620
  73 IF(T.LT.TF) GO TO 75                                                     INT14630
  74 WRITE (6,225)                                                            INT14640
 225 FORMAT (//////,26H NORMAL STOP AT FINAL TIME    )                        INT14650
     GO TO 91                                                                 INT14660
  75 IF(T.GE.TF1) GO TO 77                                                    INT14670
  76 C(13) = DT                                                               INT14680
     GO TO 87                                                                 INT14690
  77 IF(T.GE.TF2) GO TO 79                                                    INT14700
  78 C(13) = DT2                                                              INT14710
     GO TO 87                                                                 INT14720
  79 C(13) = DT3                                                              INT14730
     GO TO 87                                                                 INT14740
  80 IF(TF.GE.T) GO TO 74                                                     INT14750
```

```
      IF(TF1.LT.T) GO TO 76                                            INT14760
      IF(TF2 - T)78,79,79                                              INT14770
   87 C(10) = 5.                                                       INT14780
C                                                                      INT14790
   88 CALL RKUTTA (NN,T,X,DT,C,TC,XC,DX)                               INT14800
C                                                                      INT14810
      IF(C(10).EQ.6.) RETURN                                           INT14820
   90 T = T + DT                                                       INT14830
      GO TO 2000                                                       INT14840
   91 IF(KK.EQ.0) GO TO 330                                            INT14850
      IF(MULTIP.NE.0) GO TO 97                                         INT14860
C                                                                      INT14870
C     PRINT PLOT UP TO 4 INDIVIDUAL CURVES                             INT14880
C                                                                      INT14890
      NUMPTS=-NUMPTS                                                   INT14900
      DO 310 II=1,KK                                                   INT14910
      WRITE(6,9998)                                                    INT14920
 9998 FORMAT(1H1)                                                      INT14930
      ITITLE(9)=KTITLE(2*II-1)                                         INT14940
      ITITLE(10)=KTITLE(2*II)                                          INT14950
      GO TO (311,312,313,314),II                                       INT14960
  311 CALL PLOTP(X1,Y1,NUMPTS,0)                                       INT14970
      GO TO 310                                                        INT14980
  312 CALL PLOTP(X2,Y2,NUMPTS,0)                                       INT14990
      GO TO 310                                                        INT15000
  313 CALL PLOTP(X3,Y3,NUMPTS,0)                                       INT15010
      GO TO 310                                                        INT15020
  314 CALL PLOTP(X4,Y4,NUMPTS,0)                                       INT15030
  310 WRITE(6,9999) ITITLE                                             INT15040
 9999 FORMAT(1H0,8X,12A8)                                              INT15050
      GO TO 330                                                        INT15060
C                                                                      INT15070
C     PLOT DUMMY CURVE ALONG AXES TO SET SCALES FOR MULTIPLE PLOT      INT15080
C                                                                      INT15090
   97 BIGX = 0.                                                        INT15100
      BIGY = 0.                                                        INT15110
      SMLX = 0.                                                        INT15120
      SMLY = 0.                                                        INT15130
      DO 1970 I=1,NUMPTS                                               INT15140
      XMAX =AMAX1  ( X1(I), X2(I), X3(I), X4(I))                       INT15150
      YMAX =AMAX1  ( Y1(I), Y2(I), Y3(I), Y4(I))                       INT15160
      XMIN =AMIN1  (X1(I), X2(I), X3(I), X4(I))                        INT15170
      YMIN =AMIN1 ( Y1(I), Y2(I), Y3(I), Y4(I))                        INT15180
      IF(BIGX.LT.XMAX) BIGX=XMAX                                       INT15190
      IF(BIGY.LT.YMAX) BIGY=YMAX                                       INT15200
      IF(SMLX.GT.XMIN) SMLX=XMIN                                       INT15210
      IF(SMLY.GT.YMIN) SMLY=YMIN                                       INT15220
 1970 CONTINUE                                                         INT15230
```

```
      TX(1) = 0.                                                        INT15240
      TX(2) = 0.                                                        INT15250
      TX(3) = 0.                                                        INT15260
      TX(4) = SMLX                                                      INT15270
      TX(5) = BIGX                                                      INT15280
      TY(1) = BIGY                                                      INT15290
      TY(2) = SMLY                                                      INT15300
      TY(3) = 0.                                                        INT15310
      TY(4) = 0.                                                        INT15320
      TY(5) = 0.                                                        INT15330
      WRITE(6,9998)                                                     INT15340
      ITITLE(9) = KTITLE(1)                                             INT15350
      ITITLE(10)= KTITLE(2)                                             INT15360
      NT=-5                                                             INT15370
      CALL PLOTP(TX,TY,NT,1)                                            INT15380
      MODCUR  = 2                                                       INT15390
      DO 410 II=1,MULTIP                                                INT15400
      IF(II.EQ.MULTIP) MODCUR=3                                         INT15410
      GO TO  (411,412,413,414),II                                       INT15420
  411 CALL PLOTP(X1,Y1,NUMPTS,MODCUR)                                   INT15430
      GO TO 410                                                         INT15440
  412 CALL PLOTP(X2,Y2,NUMPTS,MODCUR)                                   INT15450
      GO TO 410                                                         INT15460
  413 CALL PLOTP(X3,Y3,NUMPTS,MODCUR)                                   INT15470
      GO TO 410                                                         INT15480
  414 CALL PLOTP(X4,Y4,NUMPTS,MODCUR)                                   INT15490
  410 CONTINUE                                                          INT15500
      WRITE(6,9999) ITITLE                                              INT15510
C                                                                       INT15520
  330 IF(NRC.NE.NR) GO TO 1000                                          INT15530
      IF(NR.GT.1) GO TO 333                                             INT15540
      WRITE(6,226)                                                      INT15550
  226 FORMAT(//,43H THE ONE RUN CALLED FOR HAS BEEN COMPLETED. ,//)     INT15560
      STOP                                                              INT15570
  333 WRITE(6,227)NR                                                    INT15580
  227 FORMAT(//,5H THE ,I1,37H RUNS CALLED FOR HAVE BEEN COMPLETED.,//) INT15590
      STOP                                                              INT15600
      END                                                               INT15610

      SUBROUTINE RKUTTA(/NN/,/T/,/X/,/DT/,/C/,/TC/,/XC/,/DX/)           INT15620
      DIMENSION X(30), C(15), XC(30), DX(30), CT(4), AK(4,30)           INT15630
      REAL*8 AK,CT                                                      INT15640
      INDIC = C(10) - 4.0+0.0000001                                     INT15650
      IF(INDIC.GT.1) GO TO 3                                            INT15660
      CT(1) = 0.0D0                                                     INT15670
      CT(2) = 0.5D0                                                     INT15680
      CT(3) = 0.5D0                                                     INT15690
```

```
      CT(4) = 1.0D0                                                     INT15700
      II=0                                                              INT15710
    7 II=II+1                                                           INT15720
      TC = T + CT(II)*DT                                                INT15730
      DO 2 J=1,NN                                                       INT15740
    2 XC(J) = X(J) + CT(II)*AK(II-1, J)                                 INT15750
      C(10) = 6.0                                                       INT15760
      RETURN                                                            INT15770
    3 DO 4 J=1,NN                                                       INT15780
    4 AK(II, J) = DT*DX(J)                                              INT15790
      IF(II.LT.4) GO TO 7                                               INT15800
      DO 5 J=1,NN                                                       INT15810
    5 X(J)=X(J)+(AK(1,J)+2.0*(AK(2,J)+AK(3,J))+AK(4,J))/6.0             INT15820
      C(10) = 7.0                                                       INT15830
      RETURN                                                            INT15840
      END                                                               INT15850
//GO.SYSIN DD *
C
C     *      *      *      *      *      *      *      *      *      *      *      *
C
THE FOLLOWING LISTING CONTAINS THE DATA CARDS AS DESCRIBED UNDER SUBROUTINE
INTEG1 USAGE. CARDS FOUR AND SIX ARE MODIFIED TO ACCEPT LARGER PARAMETER
VARIATIONS. SEE STATEMENTS FOLLOWING INT12360 AND INT12550 FOR THE NEW FORMAT.
C
C
ORGANIC DYE LASER ANALYSIS
1
 3
.000001        .000001        .001            .001            1.0
.01            30.0
100000.000.15400000018000.0000.010000000.03400000054200.000010000.0000
0000000000000000000000000000000000000000000000000000180000000000000000.0000
  TIME  CO SINGLET 1 TRIPLET 2  GROUND 3
SINGLET STATE     1    TRIPLET STATE    2    GROUND STATE    3
```

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The wavelength spectrum of a CW pumped Rhodamine 6-G organic dye laser is observed to shift approximately 150 Angstroms toward shorter wavelengths when pumped by 30 nsec pulses at a one megahertz repetition rate from a cavity-dumped Argon laser. Experimental evidence of the shift is presented, gain and rate equations are developed for a simplified dye laser model, and theoretical results are obtained for a computer simulation of the experiment. A comparison is made of the theoretical and experimental results and satisfactory

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agreement is obtained within the limits of the values of the parameters used and the assumptions made in formulating the model.

Snyder

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