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PHOTODISSOCIATION DYE LASER

Semiannual Technical Report

April 1976

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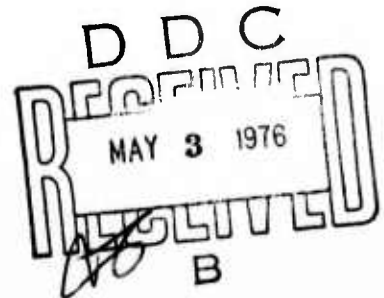
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The photodissociation dye laser (PDL) program is an effort to develop a new class of tunable liquid lasers operating in the visible region of the spectrum. In the PDL scheme, laser action is expected to occur between an excited electronic state and the ground electronic state of radicals produced by the photodissociation of specific classes of molecules in solution. This scheme is expected to result in several significant advantages over conventional dye lasers.

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During this reporting period we have completed our measurements on the potential PDL molecules that were selected from the hexaarylethane group, and have evaluated their use as active media for the PDL scheme.

Fluorescence intensity measurements as a function of excitation wavelength have been made on the hexaphenylethane-triphenylmethyl radical system. The measurements indicate that the optimum excitation source for the system is the N_2 laser. However, because the maximum gain is low ($\sim.02$ cm^{-1}), the length of active media the N_2 laser is capable of exciting is too short to overcome the single pass cavity losses. Xenon flashlamp pumping of this system has been considered, but the high intensities need (> 10 MW in the UV) makes such a pumping scheme unattractive.

In addition to hexaphenylethane, the other molecules that were synthesized in the early part of the program have been investigated. In particular, fluorescence spectra and lifetimes of pentaphenylethane, 1-p-biphenyl-1,1,2,2-tetraphenylethane, and 12-12' bifluoradenyl have been measured. These molecules have fluorescence lifetimes of ~10 ns. However, they exhibit no significant photodissociation into excited state radicals when excited in the UV (2600 Å).

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ABSTRACT

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I. INTRODUCTION

The photodissociation dye laser (PDL) program is an effort to develop a new class of tunable liquid lasers operating in the visible region of the spectrum. In the PDL scheme, laser action occurs between an excited electronic state and the ground electronic state of radicals produced by the photodissociation of specific classes of molecules in solution. In this study, a class of highly strained ethanes, the hexaarylethanes, is being investigated as potential laser media. Continuous wave operation of such lasers is expected based on the rapid relaxation of the terminal vibronic levels and the subsequent depletion of the electronic ground state by recombination to the original parent ethane structure. As in conventional dye laser systems, wavelength tunability is assured because the numerous vibrational and rotational modes of the complex molecule result in a manifold of quasi-continuous vibronic energy levels associated with each electronic level.

The photodissociation dye laser is expected to exhibit several significant advantages over conventional dye lasers. The radical can be thought of, to a first approximation, as a one-electron system. As a result, the electronic structure of the radical consists of doublet levels rather than the singlet and triplet level structure characteristic of the paired electrons of conventional dye molecules. Consequently, problems associated with non-radiative intersystem-crossing transitions which result in deleterious absorption by the lowest triplet level at the laser wavelength in conventional dye molecules are eliminated.

The second advantage of the PDL scheme is related to the upper laser level lifetime. In conventional laser dyes, the spontaneous radiative lifetime of the first excited singlet level is typically on the order of a

few nanoseconds. In radical systems, however, there is evidence, both theoretical and experimental, that the first doublet-doublet electronic transitions are in some cases, partially forbidden. From the point of view of the PDL scheme, this means that the radiative lifetime of the upper laser level can be one to two orders of magnitude longer than that exhibited by the upper laser levels of conventional organic dyes. Consequently, a larger population density in the upper level may be obtained, thereby allowing the possibility of generating higher laser output energy than can be presently attained with conventional dyes. This assumes that strong absorption from the upper level at the laser wavelength does not occur. In addition, the longer radiative lifetime favors the recombination of the radical ground state to the parent dimer at a sufficiently fast rate to permit a continuous population inversion and hence, cw laser action.

The aim of the current program is to demonstrate proof of principle of the PDL scheme. Towards that end, a number of promising chemical candidates from the hexaarylethane group of organic molecules have been synthesized and data has been obtained on those processes that are relevant to assessing their usefulness as PDL laser media.

During the present reporting period we have completed our study of the hexaphenylethane-triphenylmethyl radical system. Fluorescence intensities as a function of excitation wavelength have been measured. Results of these measurements indicate that the optimum excitation wavelength for the hexaphenylethane system is at ~3300 Å. Consequently, the N_2 laser, with output at 3371 Å is expected to be the best optical

pump. However, the low value of the maximum gain of $\sim .02$ cm^{-1} requires a longer excitation length, at fixed energy density, than realizable with an N_2 laser in order to overcome cavity losses and reach threshold. Since the hexaphenylethane fluorescence efficiency decreases as the excitation wavelength moves further towards the UV from ~3300 Å, the output of conventional flashlamp systems is poorly coupled to the excitation requirements of the hexaphenylethane molecule. Estimates show that a flashlamp system with output power in excess of 10 MW in the UV would be required to achieve .02 cm^{-1} gain over a 15 cm length, making such a pumping scheme highly unattractive.

In addition to hexaphenylethane, the other molecules that were synthesized in the early part of the program have been investigated. In particular, fluorescence spectra and lifetimes of pentaphenylethane, 1-p-biphenyl-1,1,2,2-tetraphenylethane, and 12-12' bifluoradenyl have been obtained. These systems however, exhibit no significant photo-dissociation into excited state radicals, and therefore will not fulfill the requirements of a PDL active media.

II. THE PHOTODISSOCIATION DYE LASER CONCEPT

In its most general form, the photodissociation dye laser may be understood as follows: A stable molecule, AB, in solution is optically pumped to its first excited singlet state. The absorbed pump energy exceeds the molecule's dissociation energy and two radicals are formed upon dissociation. The excess energy is partitioned as electronic and vibrational excitation in one or both of the radicals A· and B·. In particular, we consider radical A· to be electronically excited. A partial population inversion is produced between the first excited and the ground state of the radical. Laser action occurs in A· and the resulting A· and B· ground state molecules are unstable against recombination. The processes may be written as:

$$AB + h\nu_{pump} \longrightarrow (AB)^*$$

$$(AB)^* \longrightarrow A\cdot^* + B\cdot$$

$$A\cdot^* \longrightarrow A\cdot + h\nu_{laser}$$

$$A\cdot + B\cdot \longrightarrow A_2 + B_2 + AB$$
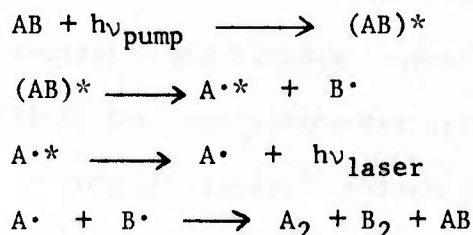
In the final recombination step, not all of the radicals form the original molecule AB, but the dimeric forms A_2 and B_2 are also produced. Continuous laser action would require replenishment of the starting material, AB. Let us consider in more detail the special case where $B \equiv A$ so that the starting molecule is a symmetrical dimer.

The general energy level scheme for such a photodissociation dye laser molecule is shown schematically in Figure 1. The stable parent dimer, having no unpaired electrons, exhibits the usual singlet and triplet level structure. The ground and excited electronic singlet levels are denoted by D_{S_o}, D_{S_1}, . . . and the triplet levels are denoted by D_{T_o}, D_{T_1}, . . .

The lowest electronic levels of the radicals, obtained from the symmetric photodissociation of the dimer, are shown adjacent to the dimer structure. The radical, having a single unpaired electron, exhibits a doublet structure. In the figure the radical ground state is shown displaced upward in energy from the dimer ground state by an amount equal to the dimer dissociation energy.

The vibrational level spacing in both the dimer and radical ranges between 150-1500 cm^{-1} while the rotational spacing ranges between 15-150 cm^{-1}. Therefore, as in conventional dye lasers, a quasicontinuum exists for each electronic level comprised of the thermally broadened rotational and vibrational levels.

A characteristic of the photodissociation dye laser molecule is that the D_{S_1} and D_{T_o} levels in the dimer lie above the dimer dissociation energy. Consequently, upon optically pumping the dimer to D_{S_1} the molecule will undergo dissociation into two radicals. Dissociation may occur via two possible paths. The first is directly from the optically pumped D_{S_1} level. However, the D_{S_1} level may, in principle, undergo a rapid intersystem crossing to the D_{T_o} level. Dissociation of the dimer may occur from this level.

Upon dissociation, the excitation energy is partitioned between a manifold of levels in the quasi-continuum of both the excited and ground state of the radical. The radical ground state initially has a negligible population; consequently, a partial inversion in the radical can be produced. The stimulated emission is tunable as in conventional dye lasers because of the quasi-continuous distribution of the upper and ground levels. An interesting point is that for radicals considered suitable for the photodissociation dye

$$A_2(D_{S_0}) + h\nu_{pump} \longrightarrow A_2(D_{S_1}, \nu'') \quad : \text{ excitation of dimer}$$

$$A_2(D_{S_1}, \nu'') \longrightarrow A_2(D_{S_0}, \nu') \quad : \text{ singlet relaxation}$$

$$A_2(D_{S_1}, \nu'') \longrightarrow A_2(D_{T_0}, \nu') \quad : \text{ intersystem crossing}$$

$$A_2(D_{S_1}, \nu'') \longrightarrow A(R_2, \nu''') + A(R_1, \nu') : \text{ dissociation}$$

$$A(R_2, \nu''') \longrightarrow A(R_1, \nu'') + h\nu_{laser} \quad : \text{ laser action}$$

$$A(R_1) + A(R_1) \longrightarrow A_2(D_{S_0}) \quad : \text{ recombination}$$
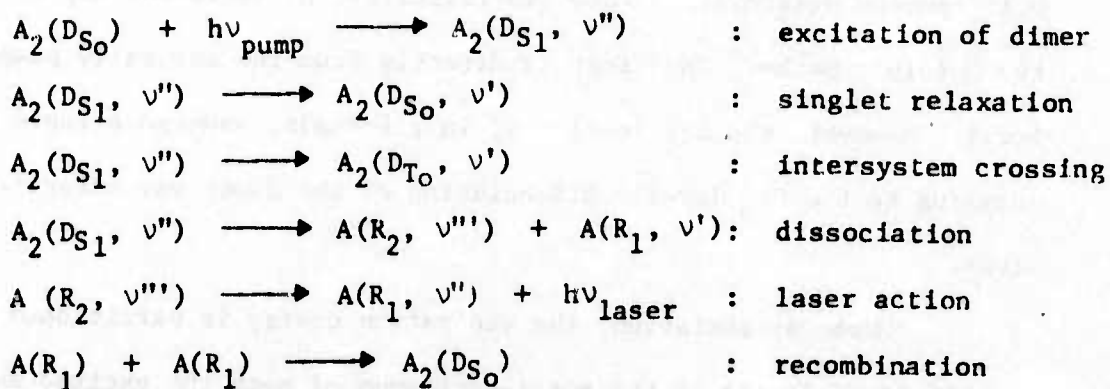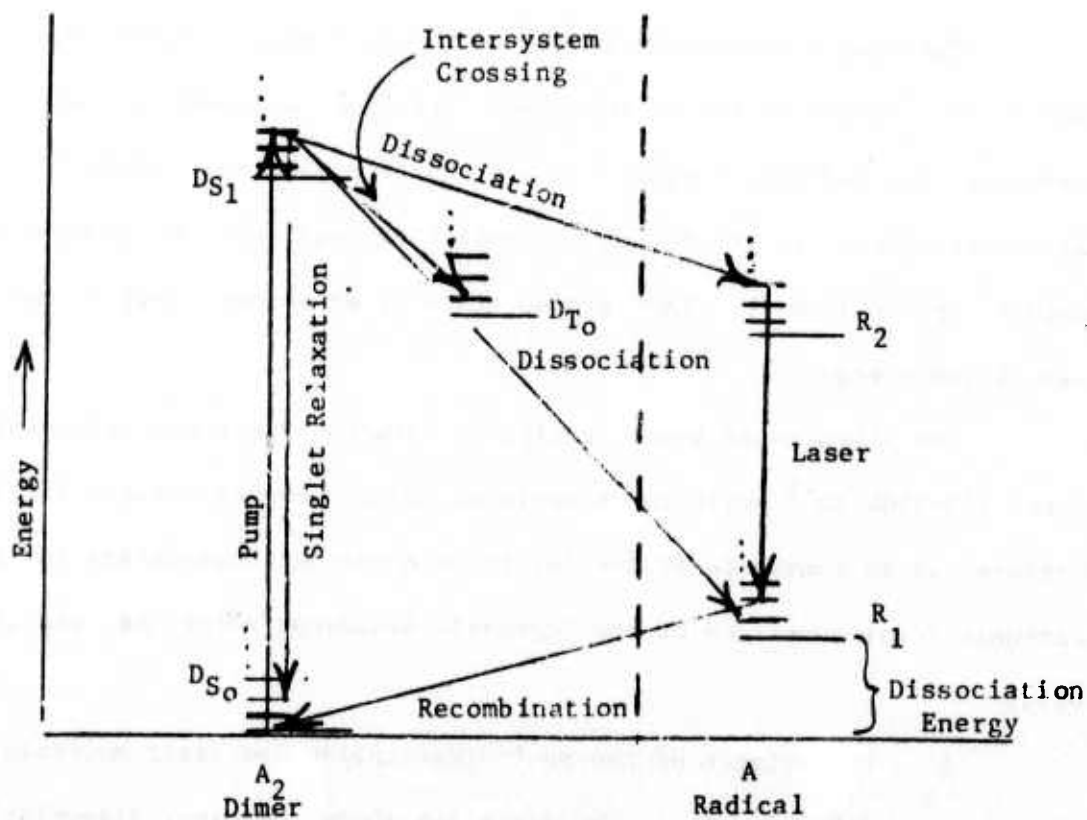
Figure 1. Photodissociation Dye Laser Molecule
Structure and Kinetics

laser both the calculated and measured oscillator strengths for the first doublet-doublet electronic transition are always small. Thus, even though these transitions are allowed by selection rule considerations, they exhibit a partial "forbiddenness". Consequently, the radiative lifetime of the laser transitions can be expected to typically be a factor of a hundred times longer than in conventional dye lasers. This has been experimentally verified in some cases as will be discussed later. The longer lived upper laser level can permit a larger population density to be obtained and thereby allows the possibility of high energy output.

The ability of the photodissociation dye laser to operate on a cw basis requires, as usual, that the depletion rate of the lower laser level exceed the radiative transition rate from the upper to the lower laser level. The lower laser level is the radical ground state which is unstable to recombination back to the original parent dimer. Thus for a steady-state population inversion to be maintained, the radical recombination rate must be greater than the laser transition radiative rate.

III. PROPERTIES OF THE SELECTED PDL MOLECULES

A class of organic molecules known as the hexaarylethanes exhibit many of the prerequisites necessary for a PDL active media. At the start of the program five compounds from this class were selected and synthesized for further study. Details on the techniques used for synthesis of these molecules were given in the first semiannual report[1].

What follows is an overview of the relevant properties of these materials known prior to our experimental effort.

A. Hexaphenylethane

The hexaphenylethane molecule can be represented schematically as:
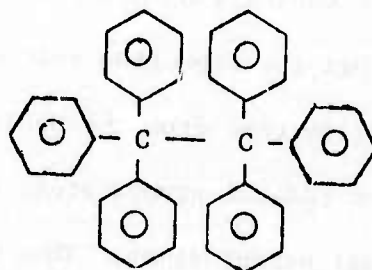
Although the accepted structure is actually non-planar, with a quinoid-type atomic arrangement, the above representation is adequate for the purposes of this discussion.

I. Chemical Properties

a. Dissociation Into Free Radicals

It has been clearly established for some time that hexaphenylethane thermally dissociates in non-reactive solvents to produce the intensely colored triphenylmethyl radical[2]:

Hexaphenylethane
(Colorless Solid)

Triphenylmethyl Radical
(Yellow in Solution)
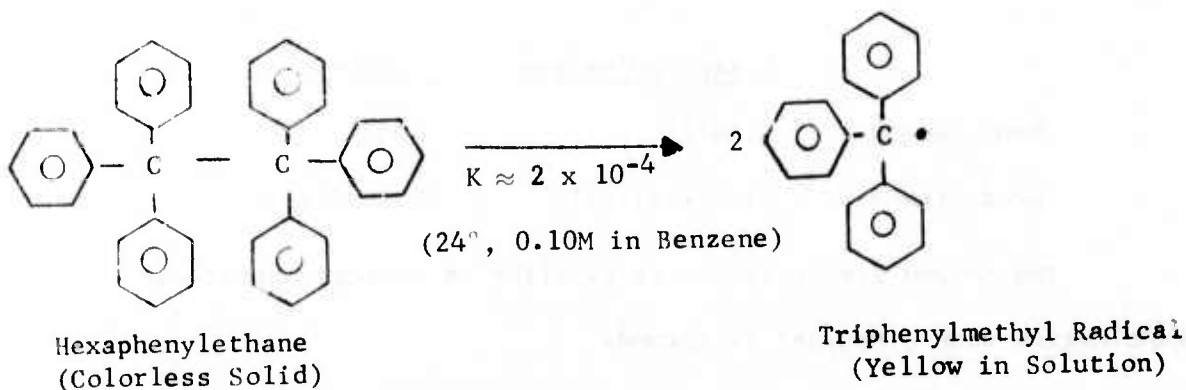
The existence of the radical has been proven by a multiplicity of analytical techniques. For example, cryoscopic molecular weight determinations of such compounds in solution have shown that the apparent molecular weights were well below that of the dimer. In addition, spectrophotometric measurements have established that solutions of the dimer do not obey Beer's law; the intensity of absorption increases with dilution as would be predicted for the dissociation of a colorless compound into a colored radical. Finally, absolute methods of radical detection - magnetic susceptibility and electron spin resonance spectroscopy - have shown beyond doubt the thermal dissociation of hexaphenyl-ethane into ground state triphenylmethyl radicals.

Two factors determine the position of the hexaphenylethane-triphenylmethyl equilibrium: (a) Steric effects and (b) Radical stability.

(a) Steric Effects - This factor favors the formation of radicals in two ways. First, there is a relief, upon dissociation, of the steric inter-actions in the ethane. In essence, the central carbon-carbon bond in the hexaphenylethane is weakened by the steric repulsion of the aromatic rings produced by interactions between the ortho-substituents. The bond weakening can be experimentally verified by comparison of the bond-length and strength of the central carbon-carbon bond in hexaphenylethane and the nonsterically-hindered ethane:

	Hexaphenylethane	Ethane
Bond Length	1.58 Å	1.54 Å
Bond Strength	11.5 kcal/mole	85 kcal/mole

The second steric effect is typified by a steric hindrance to recombination once a radical is formed.

(b) Radical Stability - The stability of the triphenylmethyl radical is provided by a resonance stabilization effect resulting from the delocalization of the free electron throughout the three aromatic rings of the structure.

b. Deleterious Reactions

(1) Disproportionation

A number of chemical properties act to complicate the use of hexaphenyl-ethane as a PDL active media candidate. These properties arise from the reactivity of the produced radicals. The first of these is their propensity towards disproportionation. This transformation is promoted by heat or light and is illustrated below:
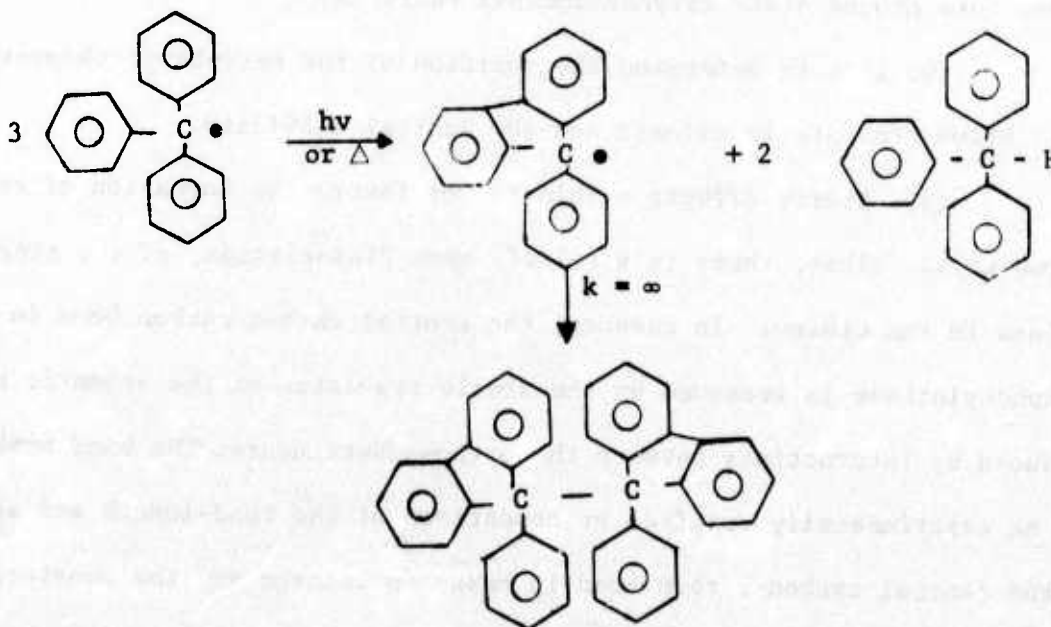
In essence, two molecules of the radical become reduced at the expense of a third radical which is oxidized. For the triphenylmethyl radical, the products are triphenylmethane and the dehydro-dimer of 9-phenylfluorene. Measurements of the effect of this photochemical or thermal degradation on the use of hexaphenylethane as a laser media were presented in the second semiannual report[3].

(2) Addition Reactions

The second deleterious reaction path which occurs when radicals are produced is the addition reaction. This involves the rapid absorption of atmospheric oxygen to form colorless triphenylmethyl peroxides:
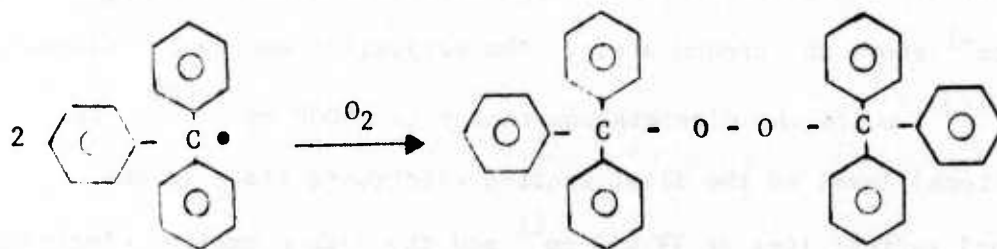
As the radicals react, more dissociation must occur in order to maintain the dimer-radical equilibrium constant. In this way, a continuous depletion of both the radical and dimer concentration occurs until all the hexaphenylethane in solution has reacted to form the peroxide precipitate. As pointed out in the first semiannual report[1], careful preparation under vacuum conditions and subsequent storage and handling under a nitrogen atmosphere yield solutions that are quite stable with respect to such reactions. Spectrophotometric measurements of the radical concentration over a period of time in solutions we have prepared indicate a decrease of only several percent per week; and this decrease is probably due primarily to thermal disproportionation.

II. Spectroscopic Properties

The reported major bands in the electronic absorption spectra of hexaphenylethane and the triphenylmethyl radical are shown below along with the measured extinction coefficients[4]:

Hexaphenylethane	Triphenylmethyl Radical
λ_{max} = 3150 Å (using KBr pellet technique)	λ_{max} = 3450 Å, ϵ = 11,000
λ_{max} = 3130 Å (dissolved in cyclohexane)	λ_{max} = 5100 Å, ϵ = 210
	(both in cyclohexane)

The major absorption bands can be associated with electronic transitions. The resulting dimer and radical energy level structure is shown in Figure 2. The first excited singlet level of the dimer, D_{S_1}, is $\sim$ 31,750 cm^{-1} above the ground state. The activation energy for dissociation is $\sim$ 7000 cm^{-1}[5] while the dissociation energy is $\sim$4000 cm^{-1} [6]. The ground vibrational level of the first excited electronic state in the triphenylmethyl radical lies at 19,410 cm^{-1} and the second excited electronic level is at $\sim$ 29,000 cm^{-1} above the ground state.

The mirror image symmetry of the absorption and fluorescence bands is illustrated in Figure 3. The spectra were originally reported by Lewis et al. [6] and were measured by suddenly cooling a solution of hexaphenylethane in EPA (5 parts ether, 5 parts isopentane, and 2 parts ethanol by volume) to liquid N_2 temperature (-190°C). It was reported that rapid cooling preserved the highly colored radicals in the clear EPA glass, and that under these conditions, there was no evidence of disproportionation. Unfortunately, no details regarding the measurement techniques used in taking the fluorescence data were presented--nor was the excitation source or wavelength described.
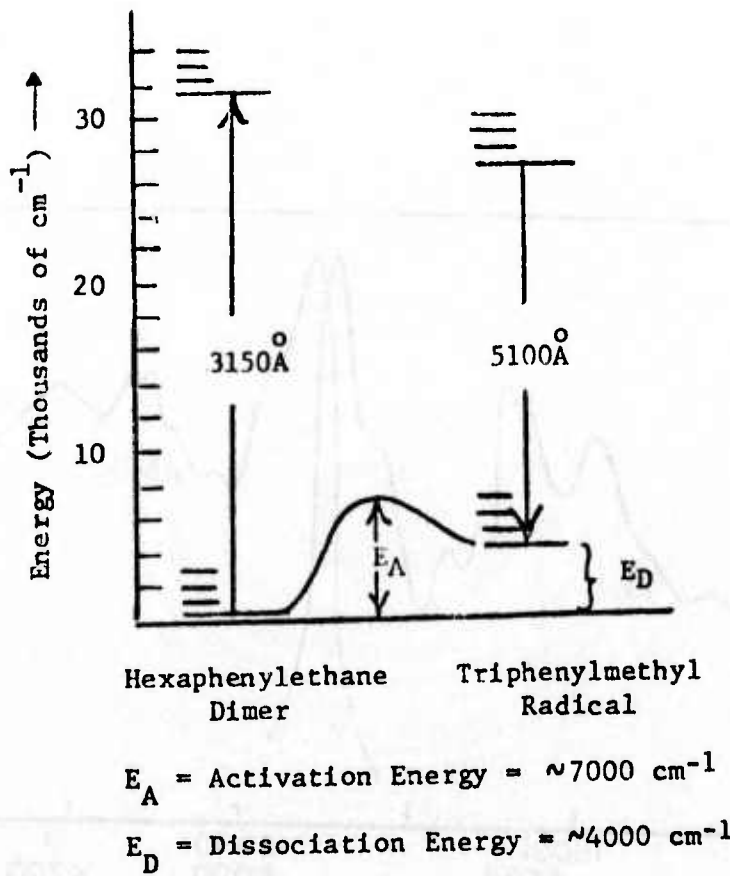
Figure 2. Energy Level Structure of the Hexaphenylethane-Triphenylmethyl System.
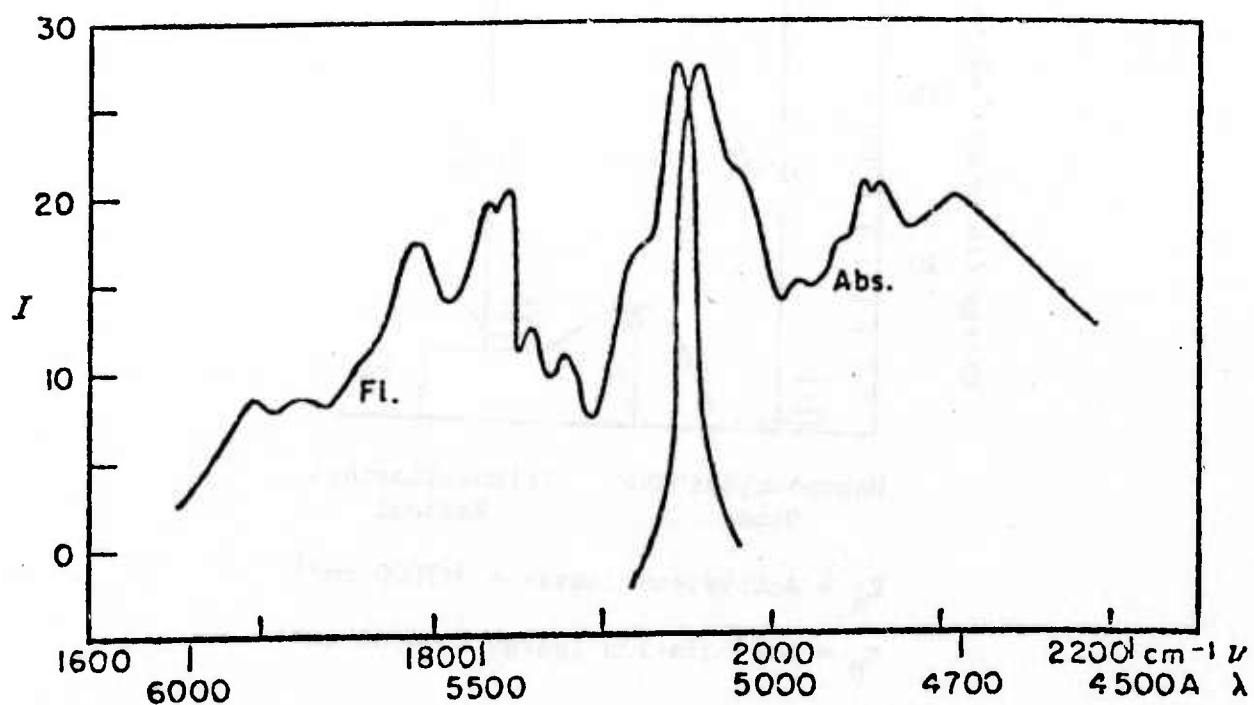
Figure 3.   Mirror Symmetry of Absorption and Fluorescence
Bands of the Triphenylmethyl Radical (in EPA
mixed solvent at -190°C). Ref. (7).

More recently, Okamura et al. [7] performed time resolved fluorescence
measurements on the triphenylmethyl and other methyl-substituted radicals
which were trapped in rigid solvents at low temperature. The triphenylmethyl
radical was prepared by the photolysis of triphenylmethane molecules at -190°C
in a quartz cell using a low pressure mercury lamp. An N_2 laser emitting
a 10 nsec, 40 kw peak power pulse at 3371 Å was used as the excitation source.
The fluorescence decay consisted of a single exponential with the measured
decay times being 280 nsec with ethyl alcohol as solvent, and 330 nsec using
isopentane as the solvent. The very long fluorescence lifetimes were taken as
evidence that the first doublet-doublet electronic transition in the radical
has a forbidden character, although such transitions are allowed by the usual
selection rule considerations. The possibility that the observed lifetimes
were actually longer than the natural lifetimes because of complex formation
of the excited state with the solvent, was ruled out by the fact that the
difference of measured lifetimes in polar and nonpolar solvents was not
appreciable.

B. Other Hexaarylethane Derivatives

The four other molecules that were synthesized have the
advantage of being completely stable at room temperature with respect
to dissociation into radicals. They all have strong absorption features
in the UV. Their structures and UV absorption data are presented
in the first semiannual report [1].

Three of the four molecules: sesquixanthydril, pentaphenyl-
ethane, and 1-p-biphenyl-1,1,2,2-tetraphenylethane are known to dissociate
into colored radicals upon heating to ~100°C, indicating the weak
bonding inherent in this group of molecules. Although no thermal or photo-
chemical data with regard to radical production is available for 12-12'
bifluoradenyl, its structure is sufficiently similar to hexaphenylethane
to warrant its inclusion as a possible PDL material.

Unfortunately, solubility problems have precluded further
study of sesquixanthydril as a potential PDL molecule. A spectroscopic
study and evaluation of the three remaining molecules is presented
in this report.

IV. EXPERIMENTAL EFFORT AND RESULTS

A. Hexaphenylethane

Aside from the synthesis of five potential PDL materials, the major part of the experimental effort reported in the previous semiannual reports has been directed towards obtaining data that is relevant to assessing the possibility of laser action in the hexaphenylethane-triphenylmethyl radical system.

On the positive side, photodissociation of the hexaphenylethane dimer into excited state triphenylmethyl radicals has been demonstrated. Optical gains of $\sim.02$ cm^{-1} have been determined from absolute intensity measurements utilizing N$_2$ laser excitation (assuming the lower laser level to be unpopulated). Fluorescence lifetimes of the first excited triphenylmethyl radical level (the upper laser level) of $\sim$200 ns have been measured at low temperature (-80°C).

On the negative side, the room temperature fluorescence lifetime is short ($\sim$15 nsec), the material must be handled in an oxygen-free environment because of the extreme reactivity of the thermally-generated radicals, and photochemical stability is poor. In addition, the effective quantum yield of fluorescence measured at the peak of the UV absorption band ($\sim$3350 Å) is low. (These measurements were performed using an N$_2$ laser whose output (3371 Å) essential coincides with the peak of the absorption band.) For every 100 3371 Å photons absorbed by the hexaphenylethane, only one photon comes out as triphenylmethyl radical fluorescence, giving an effective quantum yield of 1%. For further details on these measurements the reader is referred to the previous semiannual reports[1,3].

The first topic that has been addressed during the present reporting period is that of the dependence of the hexaphenylethane fluorescence intensity on the excitation wavelength. This study was motivated by the fact that although the peak of the hexaphenylethane-triphenylmethyl UV absorption curve lies very close to the 3371 Å nitrogen laser wavelength, the cross section for photodissociation into excited state radicals may peak at a different wavelength, and as a result, larger fluorescence yields may be obtainable at other wavelengths even though the UV absorption is less. In addition, a knowledge of how the triphenylmethyl radical fluorescence intensity varies as a function of excitation wavelength is necessary in order to make an evaluation of different possible pumping mechanisms (e.g. laser vs flashlamp).

A frequency-doubled dye laser was used to provide a tunable UV source for these measurements. The peak of the fluorescent emission (5200 Å) from the hexaphenylethane solution was passed through a mono-chromator and detected by a 1P21 photomultiplier tube. The pulsed output was time-averaged with a PAR 160 boxcar integrator. A Molectron J3-05 joulemeter was used to measure the incident laser intensity at each wavelength. Figure 4 shows the results of this series of measurements. The plotted ratio represents the hexaphenylethane fluorescence intensity normalized to the incident excitation intensity. If a comparison of this curve to the hexaphenylethane UV absorption curve (Figure 5) is made, one sees that they are quite similar in shape.

From the point of view of assessing an appropriate optical pump source for hexaphenylethane, these findings lead to some important conclusions. Optical pumping by a N_2 laser looks attractive for two reasons. First of all, the N_2 laser output of 3371 Å falls close to
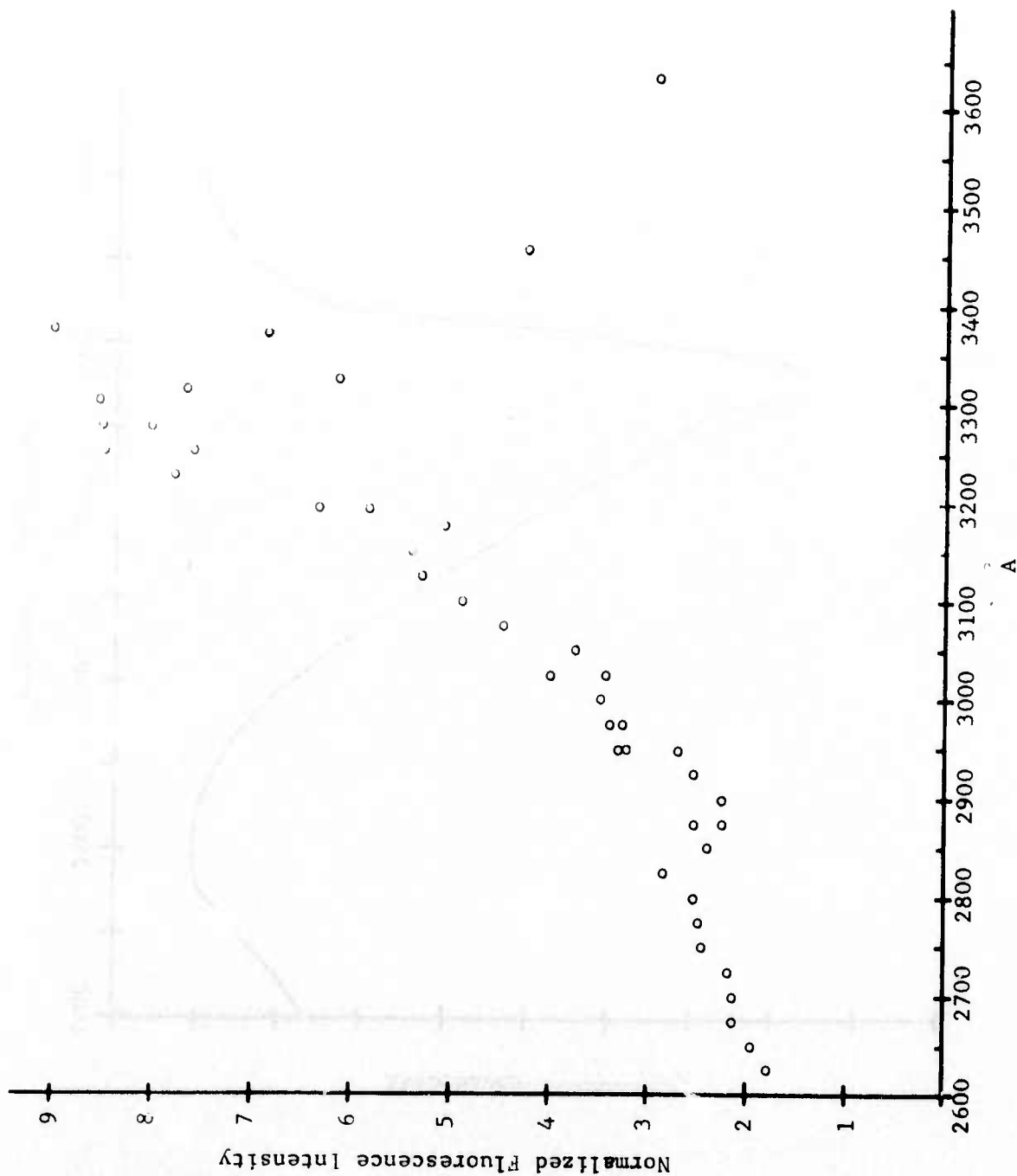
Figure 4. Hexaphenylethane fluorescence intensity plotted as a function of excitation wavelength. The fluorescence intensity shown is normalized to the incident intensity at each wavelength.
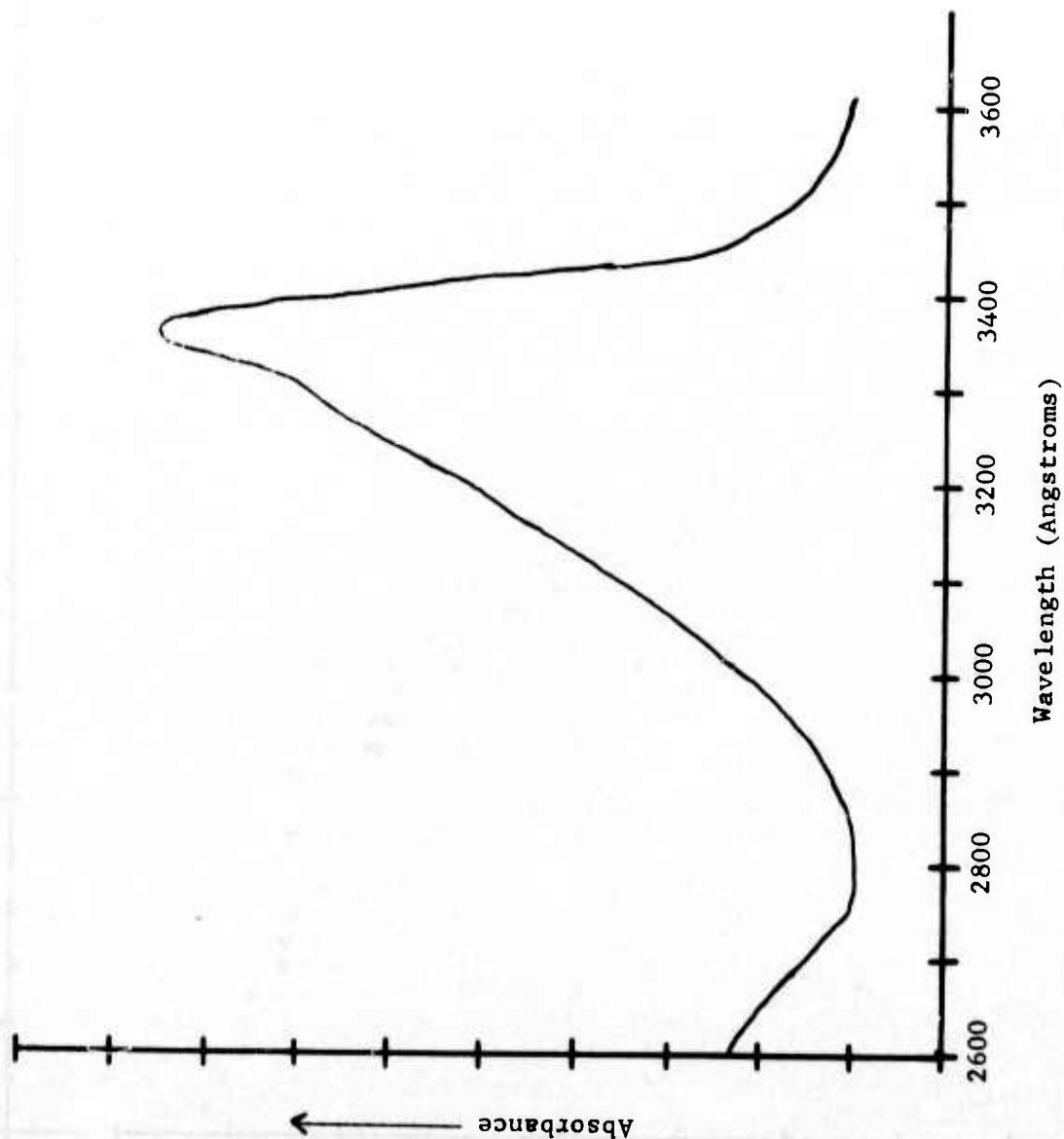
Figure 5.   UV Absorption Spectrum of Hexaphenylethane (in iso-octane).

the peak of the UV absorption curve. Secondly, optical pumping at this wavelength not only results in the maximum absorption, but in the largest fluorescence quantum yield as well.

An attempt at achieving laser action in a solution of hexaphenylethane in isooctane has been made utilizing N_2 laser pumping. The 300 kw, 8 ns output of our N_2 laser was focused onto a quartz cell equipped with brewster angle windows. Two highly reflective dielectric coated mirrors served to form a resonant cavity. However, no laser action was attained from this configuration. Unfortunately, the excitation length one can achieve with the N_2 laser for a given power density is essentially limited by the beam geometry. In our case, using a cylindrical lens to form a line image, the effective excitation length is ~2.5 cm.

Taking our estimated value for the gain coefficient of .02 cm^{-1}, this means that the single pass gain through the hexaphenylethane solution is at most 5%, and this is not sufficient to overcome other cavity losses.

Xenon flashlamp pumping has been considered as a means of exciting a longer excitation length. In the N_2 laser pumping experiment, ~10 MW/cc are absorbed by the hexaphenylethane solution. In order to maintain this absorbed power density along a 15 cm length of active region would require a flashlamp output power equivalent to ~2 MW at 3371 Å. The fact that the flashlamp output is broadband and that both the hexaphenylethane absorption and triphenylmethyl radical fluorescence falls off rapidly as one moves further into the UV from 3371 Å means that the flashlamp output must be on the order of ~10 MW in the range 2800-3600 Å in order to be equivalent to the N_2 laser pump in exciting the hexaphenylethane solution. These large pumping requirements have led us to the conclusion that Xenon flashlamp pumping of hexaphenylethane would be highly impractical.

B. Other PDL Molecules

In addition to the work on hexaphenylethane, fluorescence
spectra have been obtained during this reporting period for the other
PDL molecules that were synthesized at the start of the program. The
excitation source for all the spectra discussed below is a frequency
doubled dye laser tuned to 2600 Å. All the molecules described
below are strongly absorbing at this wavelength.

(a) Pentaphenylethane - Breaking of the central carbon-carbon
bond results in the production of both triphenylmethyl and diphenylmethyl
radicals. As mentioned previously thermal production of such radicals
has been noted historically as color changes in heated solutions of
pentaphenylethane. The fluorescence spectrum shown in Figure 6
does not show any features in the visible that can be associated with
production of these radicals in an excited electronic state. Rather,
a broad feature in the UV dominates the spectrum and is probably due
to fluorescence of the pentaphenylethane dimer structure. We conclude
that UV excitation of pentaphenylethane does not result in any significant
photolytic production of electronically excited radicals.

(b) 1-p-biphenyl-1,1,2,2-tetraphenylethane - For this molecule
as well, thermal dissociation into radicals has been observed by noting
that clear solutions turn colored upon heating due to the visible
absorption spectrum of the radicals. However, the fluorescence spectra
we have taken (Figure 7) do not show any corresponding visible fluorescence
that would be indicative of the formation of radicals in an electronically
excited state. The weak fluorescence that is observed in the UV can
be attributed to the dimer.
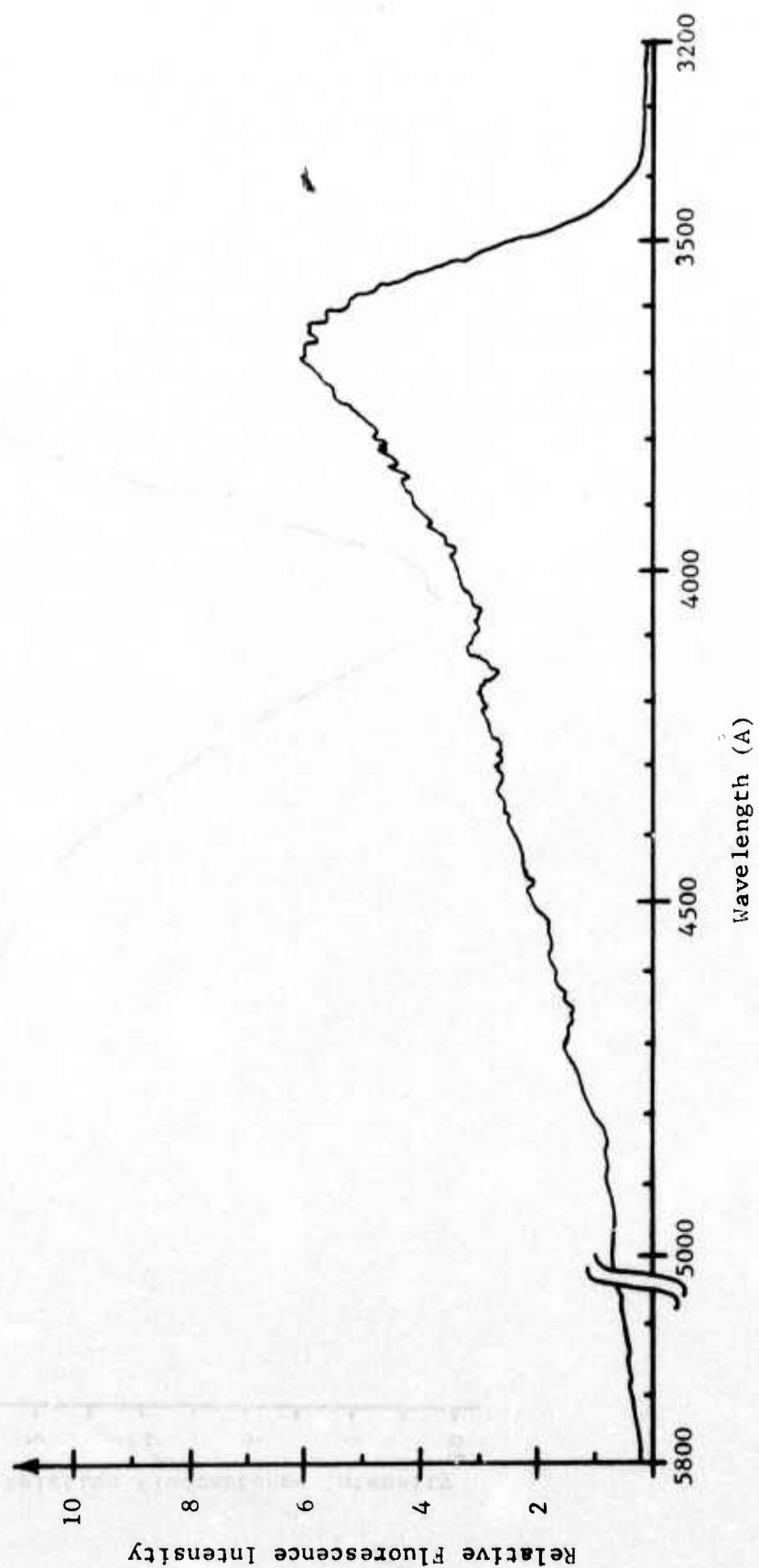
Figure 6. Fluorescence Spectrum of pentaphenylethane. The excitation source is a frequency doubled dye laser operating at 2600 Å.
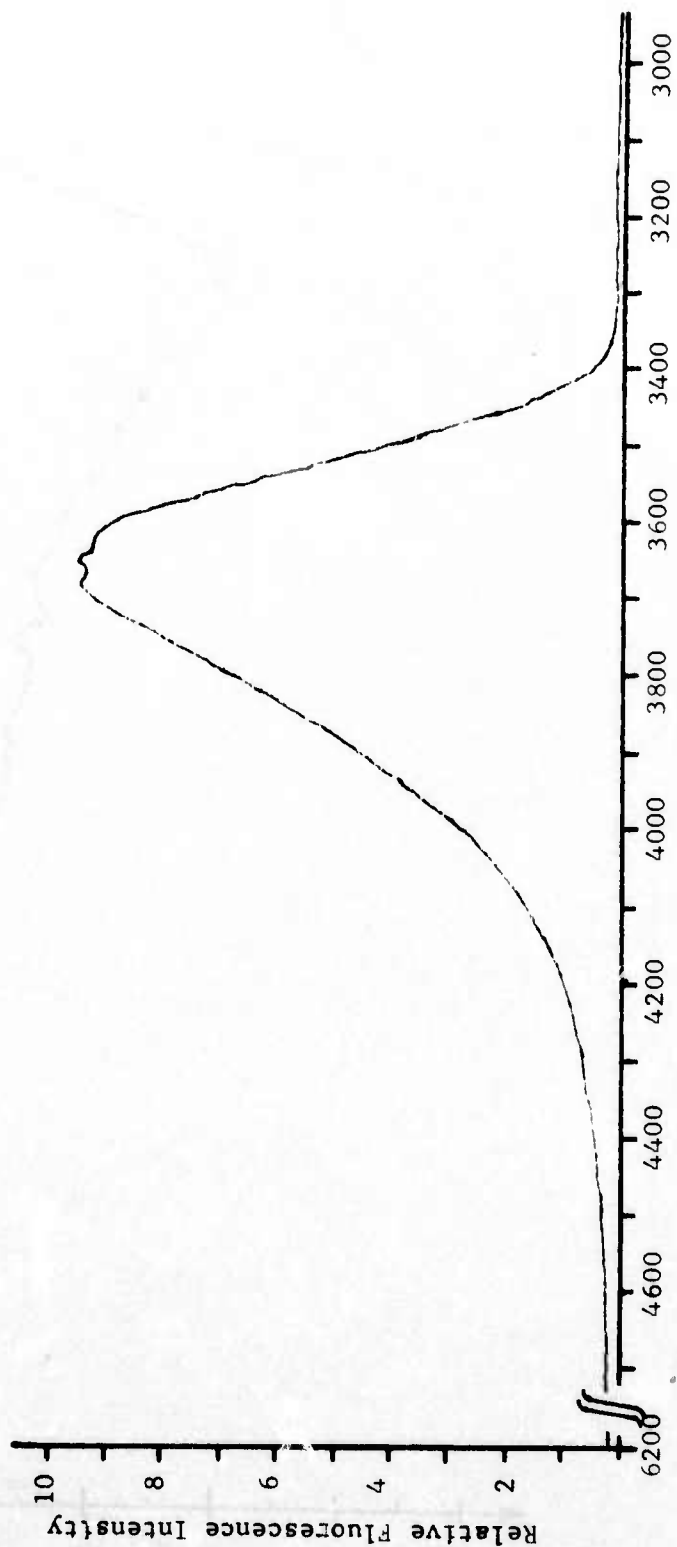
Figure 7. Fluorescence Spectrum of 1-p-biphenyl-1,1,2,2 tetraphenylethane excited by a frequency doubled dye laser operating at 2600 Å.

(c) 12-12' Bifluoradenyl - No evidence for thermal dissociation into radicals has been reported in the literature for this structure. The fluorescence spectra we have observed (Figure 8) indicate several peaks in the blue and UV. However, the fluorescence intensity is extremely weak.

The fluorescence lifetime for all these structures has been measured. The room temperature lifetimes are all ~10 ns with no observable change as the temperature of the medium is lowered to ~-100°C.

(d) We have investigated one other molecule that was not one of the original PDL molecules synthesized at the start of the program. This molecule is triphenylmethylchloride:
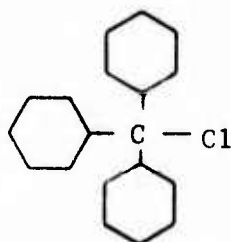
Irradiation of a solution containing this molecule with 2600 Å light results in triphenylmethyl radical fluorescence. Photodissociation resulting in excited radical states is clearly seen to occur in this material. However, the fluorescence quantum yield is less than 1%, and therefore this molecule is not suitable as a PDL active media.
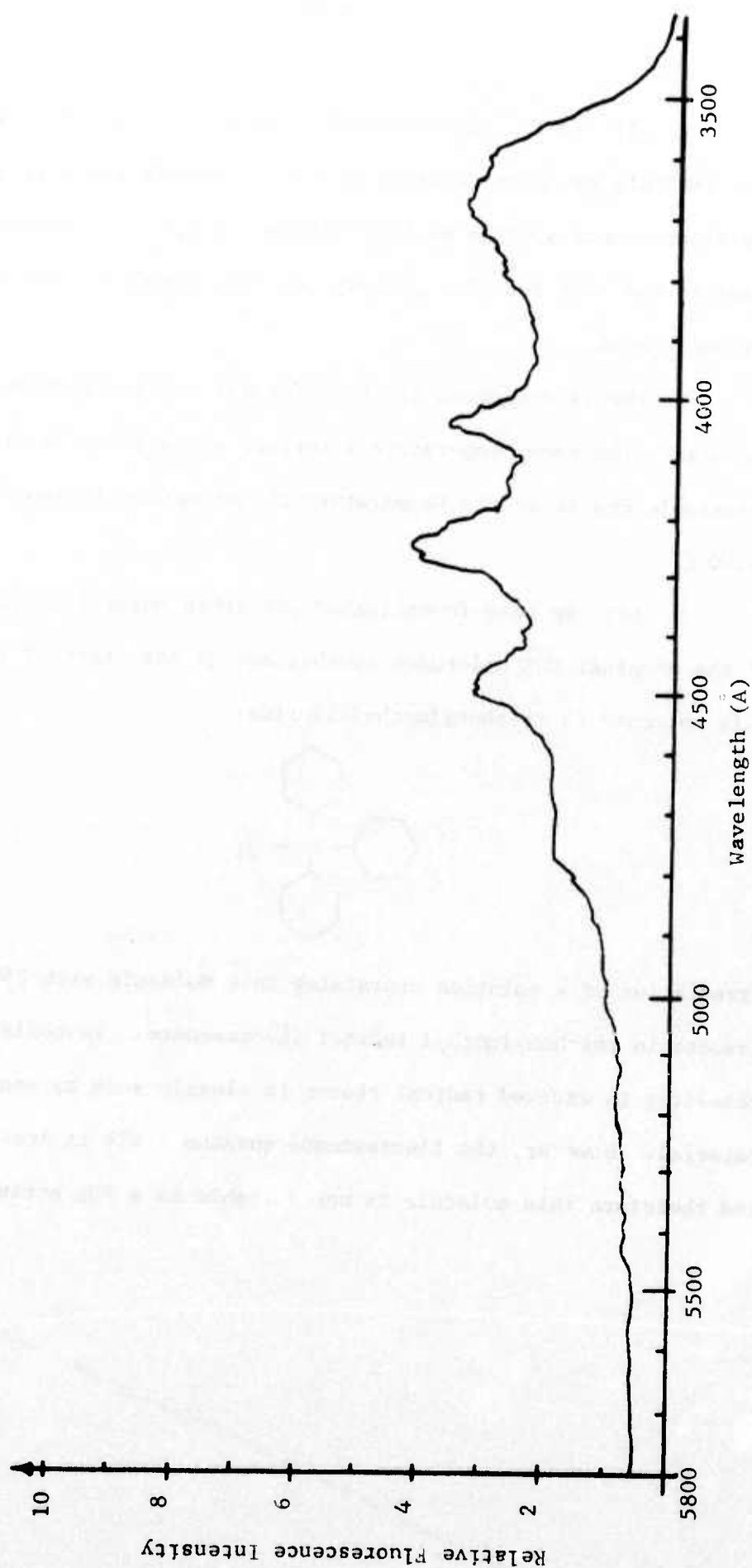
Figure 8. Fluorescence Spectrum of 12-12'bifluoradenyl excited by a frequency doubled dye laser operating at 2600 A.

V.   CONCLUSIONS

During this reporting period we have completed our measurements on the potential PDL molecules that were selected from the hexaarylethane group, and have evaluated their use as active media for the PDL scheme.

Fluorescence intensity measurements as a function of excitation wavelength have been made on the hexaphenylethane-triphenylmethyl radical system.   These measurements indicate that the optimum optical pump source for the system is the N_2 laser.   However, because the maximum gain for the system is low ($\sim.02$ cm^{-1}), the length of the active media that the N_2 laser can excite is too short to overcome the single pass cavity losses.

The remaining hexaarylethane molecules synthesized at the start of the program have been studied as well.   Their measured fluorescence lifetimes are ~ 10 ns in all cases.   Their fluorescence spectra indicate that these molecules do not undergo appreciable photodissociation upon UV excitation (2600 Å) into excited electronically radicals.   They are, therefore, not suitable as PDL active media.

Further progress in this program will involve synthesis of new candidates to fulfill the requirements of the PDL scheme.   Possible choices of new groups of organic molecules to consider are the dilactones, alkoxytetraarylethanes, and tetraphenylallyl molecules.   All these groups have been reported to reversibly dissociate into radicals as a function of temperature.

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