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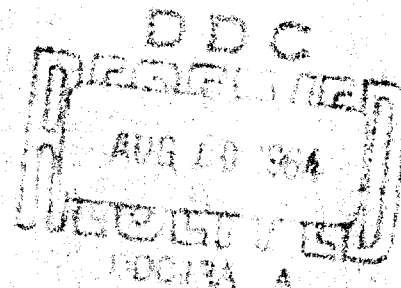
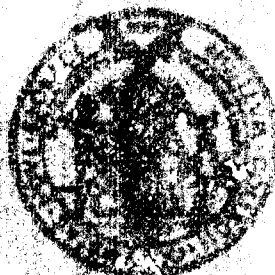
ANNUAL TECHNICAL SUMMARY REPORT

Year 268(88)

PROGRESS TOWARD A NEON-HYDROGEN LASER
and
APPLICATIONS OF OPTICAL PUMPING
FOR LASER CROSS-MODULATION

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ANNUAL TECHNICAL SUMMARY REPORT

Nonr 266(93)

INTRODUCTION

This report summarizes the significant technical results obtained under contract Nonr 266(93) between the Plasma Physics Laboratory of Columbia University and the Office of Naval Research in the last year. The material contained herein does not constitute a technical or final report of this project. A technical report (Columbia University Plasma Laboratory Report No. 8) has been distributed on the work performed in the first eight months of this project, and covers the theory and experiments of the gaseous electronics portion of the proposed Balmer- α atomic hydrogen laser project. The details and results of this work will not be repeated here.

The report is divided into two sections:

1. In the first section, certain further measurements upon the Ne-H₂ mixture are elaborated and refined, and further problems relating to the operation of a Ne-H₂ laser are discussed. The course of future investigation is outlined.
2. In the second part, we present some preliminary results of a series of experiments designed to study, demonstrate, and utilize the effect of

high power optical radiation upon an ensemble of metastables which may be contained in an operating laser. We report the details of a successful attempt to cross modulate the laser output with an external incoherent optical source and point out its implications.

I. Ne-H₂ LASER CONSIDERATIONS AND MEASUREMENTS

In this section we shall summarize the results of the gaseous electronics part of the neon-hydrogen investigation. We shall describe only the new results and techniques developed since the writing of the Columbia University Plasma Laboratory Report No. 8, "The De-Activation of Neon Metastables by H₂." The reader is referred to this¹ for all details pertaining to experimental or theoretical techniques.

The de-activation of metastables by impurity gases may occur through several processes; the most common of these is the ionization of the impurity whenever the energy of the metastable exceeds the ionized impurity state in question (Penning Effect). In our work we may measure the lifetime of the metastables, their absolute concentration, and the ionization level in the afterglow plasma which follows a weak primary discharge in the mixture. This permits a separation of the total Ne*(metastable)-H₂ de-activation cross section from the partial ionizing cross section. Total cross sections for de-activation at 300°K gas temperature in a low-pressure, low electron-density afterglow ($p \sim 1$ mm Hg, $n_e \lesssim 10^{10} \text{ cm}^{-3}$) are given in abbreviated form in Table I. We wish to emphasize two points about these results with regard to a possible Neon-Hydrogen laser. operating at the 6563 Å wavelength:

1. To form the $n = 3$ excited state of atomic hydrogen, the H₂ molecule must be dissociated:

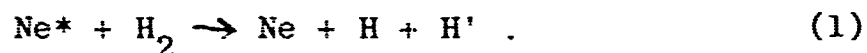


TABLE I

<u>Reaction</u>	<u>Total metastable- destruction σ (cm²)</u>
Ne*(³ P ₂ , 1S ₅) + H ₂	0.7 x 10 ⁻¹⁵
Ne*(³ P ₂ , 1S ₅) + H ₂ (T = 650°K)	2.5 x 10 ⁻¹⁵
Ne*(³ P ₂ , 1S ₅) + D ₂	1.1 x 10 ⁻¹⁵
Ne* + CH ₄	2.5 x 10 ⁻¹⁵
Ne* + NH ₃	8 x 10 ⁻¹⁵
He*(³ S ₁) + H ₂	2.5 x 10 ⁻¹⁶

When the excited hydrogen atom detaches, it will emit doppler-broadened radiation after a time considerably greater than that required to escape from the force field of the excited collision complex ($\sim 10^{-13}$ sec.). We therefore expect the Balmer α ($H\alpha$) radiation to be characteristic of an essentially unperturbed moving radiator. Since hydrogen is not massive, it is essential that the energy defect in the dissociation reaction be $\lesssim kT$ to limit the doppler broadening; this will also insure that the probability of the energy transfer is appreciable. The energy surplus in creating $H + H(n = 3) + Ne$ is indeed quite small in $Ne^* - H_2$ or D_2 collisions, and $H\alpha$ fluorescent light was observed to follow the decay of the Neon metastable density in the early ($t \lesssim 100 \mu$ sec.) afterglow. No other spectral line radiations were observed at this time, although it is clear that $L\alpha$, $L\beta$ must be emitted if $H\alpha$ is.

The hydrogen bonding energy does not vary greatly from one molecule to another and hence one might expect a similar dissociative-transfer to occur for other simple gaseous hydrogenic molecules such as CH_4 or NH_3 . Exactly the same effects were observed: the neon metastables were rapidly quenched by the impurity gas, while $H\alpha$ light was emitted. Experiments were conducted on a single-shot pulsed basis, since it was by no means evident that CH_4 or NH_3 will be stable against reaction in a gaseous discharge. Since the hydrogen bonding energy of these molecules is not appreciably different from that of H_2 , the free excited hydrogen atom ejected

from CH_4 or NH_3 should be moving much more slowly than in the case when H_2 was the parent. The oscillation threshold condition of the $\text{H}\alpha$ laser would then be less rigorous.

2. Given a suitably large cross section for de-activation, one must next inquire into the fraction of such collisions which yield an excited hydrogen atom in the $n = 3$ state. Providing the energy balance of the reaction is good, serious competition comes from ionization only, since ionization releases a free electron which may carry off the internal energy defect with much higher probability than a heavy particle. The Helium-Hydrogen collision studied above is an example. The triplet metastable energy of Helium is 19.8 eV which may excite many modes in H_2 ; however, we expect ionization ($\text{H}_2^+ + e^-$) to dominate. No $\text{H}\alpha$ radiation was observed in the afterglow of a He- H_2 mixture as might be anticipated.

Initially our measurements of the ionization cross section (σ_i) for the $\text{Ne}^* - \text{H}_2$ collision were only partly successful in view of the rather low electron densities to be determined. From Figure 1, one observes that the energy of the neon metastable (16.6 eV) is sufficient to form only $\text{H}_2^+ + e^-$ of all possible hydrogenic ionizations. Formation of NeH^+ has not been detected by others²; generation of H_3^+ does not create additional electrons. When the gas discharge is switched off, the electron density may be shown to decrease according to

$$n_e(t) = e^{-t/\tau_e} \left[n_{e0} + \frac{N_{m0} \nu_i \tau_e \tau_m}{\tau_e - \tau_m} \right] - \left[\frac{N_{m0} \nu_i \tau_e \tau_m}{\tau_e - \tau_m} \right] e^{-t/\tau_m} \quad (2)$$

where n_{e0} , N_{m0} are the measured electron and metastable densities at $t = 0$; τ_e , τ_m are the measured decay constants of the diffusing electrons and metastables, and ν_i is the ionization rate. The primary interpretational difficulty in this experiment is the determination of N_{m0} , since the absolute metastable density depends on the relatively uncertain value of the neon line oscillator strengths. We found that the ionization rate in the $\text{Ne}^* - \text{H}_2$ collision was much smaller than in the example of the Penning reaction ($\text{Ne}^* - \text{A}$), or $\text{He}^* - \text{H}_2$ for that matter.

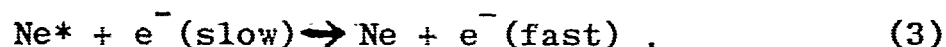
Recently, we improved the precision of this experiment considerably by measuring n_e with the plasma-post in a waveguide technique,³ reducing the frequency of the probing microwaves to a value near the guide cutoff. The resulting increase of the guide wavelength permitted a much more sensitive determination of n_e . A microwave frequency of 3.5 Gc in standard C-band waveguide was used. The graph (Figure 2) shows the effect of ionization of H_2 by Ne^* in the early afterglow. The $\text{H}\alpha$ intensity or the metastable self-absorption of neon probing radiation at $5888\text{\AA}^0$ provided separately determined exponential metastable decay rates (τ_m) in good agreement: τ_E was determined by examining the slope of the $n_e(t)$ graph at a time when few metastables were present. The σ_i obtained from this experiment was still, however, in very good agreement with that obtained many months earlier in different apparatus and reported previously,¹ $\sigma_i = 0.6 \times 10^{-16} \text{ cm}^2$.

Equation (1) might be criticized for over-simplification, since it assumes that the electron decay rate is a constant in the early afterglow. This was, however, found to be an accurate approximation for pure neon afterglow when $n_e \lesssim 10^{10}/\text{cc}$, and we are justified in using it here, providing the cooling rate of electrons is not appreciably changed by addition of H_2 . For the pure gas, the time constant for elastic cooling of warm electrons at 1 mm neon pressure is $\sim 300 \mu\text{sec}$, and hence one expects the electron temperature in the early ($t \lesssim 100 \text{ sec}$) afterglow to be relatively constant. Addition of $\frac{1}{4}\% \text{H}_2$ may be shown to increase the electron cooling rate $\sim 10\%$.⁴ However, we also note that the electrons liberated by the ionizing reaction are energetic ($\sim 1 \text{ ev}$), and that these would tend to maintain the early afterglow isothermal with a relatively constant diffusion frequency.

The conclusions of this experimental series are therefore that (1) the de-activation rate of Ne metastables by H_2 is appreciable, (2) ionization effects will not offer serious competition to modes of de-activation involving neutral particles, and (3) of these, the mode of decay resulting on one excited hydrogen atom in the $n = 3$ Bohr state would be expected to dominate, since it offers the best energy balance. Some evidence for this is provided by the measurement of the reaction rate at elevated temperatures. The rapid, observed increase of σ (Table I) implies an activation energy which is characteristic of nearly-resonant reactions, and which provides further support for the

secondary importance of σ_1 . We might therefore expect that operating the $\text{Ne}^* - \text{H}_2$ system at $\sim 200^\circ\text{C}$ would improve the fraction transferred into the channel yielding the $\text{H}\alpha$ light. As a further technical application, we expect a gaseous discharge in a $\text{Ne} - \text{H}_2$ 1% mixture (for example) to be a reliable source of ultraviolet Lyman α, β and of atomic hydrogen.

Evidence was presented that the $\text{H}\alpha$ output of low intensity gaseous discharges ($\lesssim 30 \text{ ma/cm}^2$) in $\text{Ne} - \text{H}_2$ mixtures produced an excessive amount of $\text{H}\alpha$ compared with $\text{H}\beta, \gamma$, etc. In pure neon, the metastable density was observed to saturate with $N_m \lesssim 2 \times 10^{11}/\text{cm}^3$ as $n_e \rightarrow 10^{11}/\text{cm}^3$. Addition of H_2 to such a discharge caused an expected decrease in Ne^* density, but simultaneously relaxed the saturation effect. This may be understood if we hypothesize that the hot free electrons in the discharge tube remove the metastables by a reaction of the form



Let $\alpha_m n_e N_m$ be the rate of metastable creation by electron bombardment under the given discharge conditions including cascading from upper states, and $M [D_m/\Lambda_m^2 + \alpha_E n_e + \alpha_H(\text{H}_2)]$ represent the loss rate of metastables by diffusion, electron collisions, and contact with the H_2 impurity. In the pure gas saturation obtains when $\alpha_e n_e \gg D_m/\Lambda_m^2$, where Λ_m is the diffusion length of the container, since the limiting metastable density becomes independent of n_e . However, when the impurity H_2 is introduced, saturation results at

the higher electron concentration

$$\alpha_e n_e \gg D_m / \Lambda_m^2 + \alpha_H(H_2) . \quad (4)$$

Addition of trace amounts of H_2 does not alter the mean electron energy measurably, but has been found (by a probe study) to deplete the high energy distribution ($E_e \gtrsim 6$ ev). This is quite reasonable in view of the lower ionization potential of H_2 (15.6 ev) and its many levels which may be excited when the electronic energy < 16.6 ev. On the other hand, we also observe that removing neon metastables will reduce the probability of ionization of neon by cumulative excitation. That is, instead of volume ionization via



we have as well $Ne^* + e^-(> 5 \text{ ev}) \rightarrow Ne^+ + 2e^-$. Thus, the removal of metastables should alter the ionizing efficiency of electrons in the discharge in a complex manner.

A consequence of the above is that we expect the de-activation rate of neon metastables by H_2 to maximize. Were saturation not relieved by addition of H_2 , the de-activation rate would be otherwise constant for $\alpha_H(H_2) \gtrsim D_m / \Lambda_m^2$. In Figure 3 we see that such a maximization does occur when the D_2 concentration (used here to reduce the $H\alpha$ doppler broadening) becomes $\sim 1\%$ of the neon. If the gas temperature of the mixture were to change, thereby altering the de-activation cross section, one would expect a corresponding shift in the maximum to a lower impurity

concentration. Fortunately, for a laser study, the concentration of H_2 or D_2 at which this occurs is not high, and the problem associated with a large quantity of dissociated hydrogen which would self-absorb the $L\alpha$ transition needed to empty the lower laser level ($n = 2$) does not arise. In Appendix I of Ref. (1), it was shown that the resonance-radiation trapping increase of lifetime of the $n = 2$ state would not be appreciable for generation rate of H of $< 10^{17}/\text{cm}^3\text{-sec.}$, ($3 \times 10^{13} \text{ cm}^{-3}$ of H tolerable, or $\sim 5\%$ dissociation).

Further experimentation strongly recommends looking for oscillation at 6563A in the discharge afterglow following a pulse $\sim 50 \text{ ma/cm}^2 \times 50 \mu \text{ sec.}$ First, it was found that the $H_{\alpha,\beta}$ light output grows continually during the length of the pulse, whereas the neon metastable density maximizes at $\sim 50 \mu \text{ sec.}$ Thus the electronic bombardment in the discharge is causing (H) to accumulate rapidly and, since the recombination time of H via the walls is at least $\sim 200 \mu \text{ sec.}$ ⁵, this atomic concentration will persist in the afterglow whereas the metastables will de-activate in a time only $\sim 50 \mu \text{ sec.}$ following termination of the breakdown pulse. The creation of H by metastable-de-activation alone is not serious in the afterglow; however, electronic H_2 -dissociation in an intense, continuous discharge would be detrimental to operation of the $H \alpha$ laser. (In such discharges it is not unusual to find 30% of the H_2 dissociated.⁶) Secondly, both the $n = 3$ and $n = 2$ states are radiatively connected to the ground state, which implies that in the presence of a plasma of high energy

electrons n_3/n_2 will approach a Boltzmann ratio. This follows because the electronic excitation rate depends to first order upon the dipole matrix element squared (Born Approximation) as does the radiative transition probability. Under these circumstances, the presence of free energetic electrons will tend to undo the population inversion necessary for oscillation.

Turning our attention to the early afterglow where the above factors will not be important, one calculates that for the $3^2D_{5/2} \rightarrow 2^2P_{3/2}$ hydrogen transition, assuming a population in the lower state $\sim 10\%$ of n_3 , that about 2×10^7 particles/cm³ must be accumulated in the $D_{5/2}$ level in a laser tube ~ 200 cm long with end losses of 1%. The $3^2D_{5/2}$ state will probably not receive more than the statistical weight $1/3$ of all the particles populating n_3 , which, because of ionization and competing reactions, could not itself gather more than $\frac{1}{2}$ of the total $Ne^* - H_2$ de-activation. These conditions therefore require a Ne^* de-activation rate $\gtrsim 6 \times 10^{15}$ /cm³-sec., which appears to be within reach even after diffusion losses and the lower metastable population in the afterglow are permitted. We are therefore presently assembling a 3.6 meter afterglow-type laser tube which may be pulsed with the quoted current specifications and which may be heated externally. Gas mixtures of $Ne + D_2$, CH_4 , and/or NH_3 will be studied.

The $H\alpha$ "doublet" is actually composed of several overlapping transitions (Figure 4). Depopulation of the n_3 state will cause an accumulation of particles in the

atomic hydrogen metastable level $2^2S_{1/2}$, which is separated from the peak of the desired lasing transition by about three doppler line widths. It is therefore conceivable that the H^* would introduce enough loss into a very long laser tube to prevent oscillation at the $3^2D_{5/2} \rightarrow 2^2P_{3/2}$ wavelength. The other n_2 states, $2^2P_{1/2, 3/2}$, decay radiatively in a time $\sim 10^{-9}$ sec. Given the photon cross section of the $2^2S_{1/2} \rightarrow 3^2P_{3/2}$ transition of $\sim 10^{-12} \text{ cm}^2$, and that this absorption at the peak of the $3^2D_{5/2} \rightarrow 2^2P_{3/2}$ transition is $\sim 10^{-2}$ of the maximum (conservatively two doppler width separations for gaussian lines*), then the laser photon mean free path is $\Lambda_p \sim 10^{14}/N_{2S}$ where N_{2S} is the metastable density. For the absorption to be 0.1% over a length $L > 500 \text{ cm}$, we require $L/\Lambda_p < 10^{-3}$, or

$$N_{2S} \lesssim 10^8/\text{cm}.$$

A calculation is therefore in order to estimate the lifetime of the $2^2S_{1/2}$ metastables in the early afterglow.

In the afterglow, the metastable hydrogen $2S$ atoms move in an electric field of thermal free electrons and ions. Bethe and Salpeter⁷ give the lifetime of the $2S$ state perturbed by an electric field as $(E/E_0)^{-2} t_p$, $E_0 = 475 \text{ v/cm}$ and $t_p =$ lifetime of the $2p$ state ($1.6 \times 10^{-9} \text{ sec.}$).

* The hyperfine splitting even in the $n = 2$ states is $\sim 50 \text{ mc}$, and may be neglected as a line-broadening mechanism. (Ref. 7, p. 110)

We estimate the transition probability $W_{2S} = (E/E_0)^2 W_{2p}$ from charge collisions as follows. The orbit of the charge passing the neutral metastable will be essentially a straight line with impact parameter b ; we define $r^2 = z^2 + b^2$ and take the instantaneous field strength at the 2S metastable as $\approx e/r^2$. The total probability for $2S \rightarrow 1S$ transition during the collision is

$$\bar{W}_{2S}(b) = \int_{-\infty}^{\infty} W_{2S}(t, b) dt = \frac{W_{2p}}{E_0^2} \int_{-\infty}^{\infty} \frac{e^2}{r(t, b)^4} dt. \quad (6)$$

changing the time variable with $z = vt$,

$$\bar{W}_{2S}(b) = \frac{W_{2p}}{E_0^2} \frac{e^2}{v} \int_{-\infty}^{\infty} \frac{dz}{(z^2 + b^2)^2} = \frac{\pi}{2} \frac{W_{2p} e^2}{E_0^2 v b^3}. \quad (7)$$

Since the collisions occur in a space-charge neutralized plasma, we may introduce an upper bound to the impact parameter, the Debye radius. The latter is $\sim 10^{-3}$ cm, and is clearly much larger than the minimum impact parameter which should be roughly of atomic scale (at this distance the atomic charge cloud screens the simple coulomb potential with a polarization correction). The fraction of electrons having impact parameter b in the range db is $2bdb/b_{\max}^2$. Hence,

$$\langle \bar{W}_{2S} \rangle = \frac{\pi W_{2p} e^2}{E_0^2 v b_{\max}^2} \int_{b_{\min}}^{b_{\max}} \frac{bdb}{b^3} \approx \frac{\pi W_{2p} e^2}{E_0^2 v b_{\max}^2 b_{\min}}. \quad (8)$$

The total number of electrons hitting a platelet of area $\pi b_{\max}^2$ /sec. is $n_e \pi b_{\max}^2 v$, so the transition probability per second of a field-induced $2S \rightarrow 1S$ transition is

$$w_{2S}/w_{2p} \approx \frac{\pi^2 e^2 n_e}{E_o^2 b_{\min}^2} \quad (9)$$

A typical average electronic energy in the early afterglow ($n_e \sim 10^{10} \text{ cm}^{-3}$) would be $\frac{3kT}{2} \sim \frac{1}{10} \text{ ev}$. The closest such a thermal electron may approach the atom is a distance $\sim \hbar/2\sqrt{3mkT} \sim 3\text{\AA}$, which is rather larger than the hydrogen Bohr radius ($\sim 1/2 \text{\AA}$). We compute $w_{2S}/w_{2p} \approx 1$ for these conditions, even neglecting ion or neutral particle collisions. The density of the $2S$ metastables must therefore be no greater than $10^7/\text{cm}^3$, and clearly presents no capability for absorbing the laser radiation. The calculation suggests that all levels in the $n = 3,^8$ or $n = 2$ states are stark-mixed even in this low density afterglow. A major loss to the $n = 3$ state is therefore the emission of $L\beta$, which should (for negligible self-absorption) carry off almost $2/3$ of the particles from this state.

II. OPTICAL PUMPING OF Ne* AND ITS GAS LASER IMPLICATIONS

(with B. Pariser)

For gaseous discharges or afterglows occurring in low pressure neon we have demonstrated the existence of a sizeable neon metastable $1S_5(^3P_2)$ population by optical absorption techniques. This metastable state occurs in the $1S^2 2S^2 2p^5 3S$ configuration of neon (Figure 1), two sub-levels of which may decay radiatively, emitting self-absorbed ultraviolet radiation which should have a net lifetime⁹ of $\sim 30 \mu$ sec. in a container of the type we use. The lowest state is properly metastable, with a diffusion lifetime $\sim 300 \mu$ sec. in pure Ne gas at $300^\circ K$, 1mm Hg pressure. The populations of the next two lower levels in this configuration are also appreciable ($\sim 10\%$ of $1S_5$) owing to metastable-neutral collisions which exchange an energy $\sim k T_{\text{gas}}$ required for excitation of these states from the $1S_5$. During and after the gaseous discharge, particles accumulate in $1S_5$ by cascade via the $2p^5 3p$ states. The Laporte rule (parity change) forbids direct decay from the latter configuration to the ground state ($2p^6$).

In the operation of the typical He-Ne gas laser, the particles are deposited in the $2p^5 3p$ states by transitions from the $2p^5 4S$ or $5S$ levels. It is therefore clear that the excitation of the lower laser level must pass away via the metastable bottleneck, and that accumulation of particles in the $2p^5 3S$ and $3p$ states will

limit the power output of the gaseous laser. Our object is to show how this problem may be handled radiatively, and how an external lamp may be used to control laser power output and possibly to increase it. For simplicity, the calculations and initial experimental work will be performed for the afterglow-type plasma, where the electronic energy is $\sim k T_{\text{gas}}$ and the fraction of ionization is sufficiently low that electron collisional effects will be negligible.

To begin, suppose we attempt to conduct the following experiment (Figure 5). Imagine an afterglow plasma contained in a tube placed on the axis of an optical cavity. Through this tube may be passed an axial beam of $6402\text{\AA}$ ($1S_5 \leftrightarrow 2p_9$) doppler-broadened probing neon radiation obtained from a capillary lamp. Owing to the $1S_5$ absorption, this beam will be attenuated by the decaying $1S_5$ concentration in the tube. In the course of the gradually decreasing afterglow, the ensemble of metastable atoms will be exposed to the radiation of a Xe flash tube, which provides a pulse of light having a radiation temperature $\gtrsim 5000^\circ\text{K}$ black-body equivalent in the red-orange spectral region for $\sim 50 - 100\mu\text{sec}$. At the time of the flash, we have determined that the contents of the plasma are $\sim 10^9$ electrons/cc, $\sim 10^{10} - 10^{11}$ $1S_5$ states/cc, and $\lesssim 10^{10}$ $1S_4$ or $1S_3$ states/cc. The decay rates of these states may, of course, be determined in the absence of the flash, and comprise the rate of de-activation of each $2p^5 3S$ configurational state from all processes in the afterglow. All other upper neon levels will be empty, since these should depopulate in a time not exceeding the cooling rate

of the hot electrons ($\sim 10 \mu$ sec. for 1 mm Ne). No ultraviolet radiation having $\lambda \lesssim 3000\text{\AA}$ will be present inside the neon tube when the Xe lamp is flashed; furthermore, there will be available to us Xe-lamp filters in the visible and infrared of the following type:

<u>Type</u>	<u>Pass-band</u>
IR transmitting gelatin	$2\mu > \lambda > .7\mu$
IR filtering glass	$.3\mu < \lambda < .7\mu$
gelatin	$.3\mu < \lambda < .5\mu$

The energy in a given spectral band at the discharge tube location may be determined in two steps:

1. Calibration of the Xe flash lamp outside the cavity against a corrected Tungsten strip filament standard lamp, to yield the surface brightness of the flash.
2. Comparison of the radiation sampled inside the (unfilled) discharge tube from the Xe lamp against the radiation received at the lamp's surface, the apparatus having been assembled in the cavity. The light may be conducted from the cavity to the photocell via a fibre-optic light pipe and a reflecting prism.

In Figure 6, we see the influence of the Xe flash upon the metastable $1S_5$ concentration. Following the flash, there is a quenching $\sim 25\%$. While the flash is

on, observation of the $1S_5$ density was complicated by light leakage from the flash-lamp at the $6402\text{\AA}$ probing wavelength; this could, however, be subtracted, and we notice that during this flash the neon $1S_5$ density declined to $\sim 30\%$ of that obtained just prior to the flash.

Physically, we interpret this as follows. While the flash is on, an appreciable population accumulates in the upper neon levels from radiative pumping, particularly in the ten states of the $2p^5 3p$ configuration. However, because neon is not strictly speaking a Russell-Saunders atom, when a particular level in the $3p$ configuration decays to the $3S$ it may not return to the $1S_5$ state, but rather to another state having a shorter lifetime. Thus, the population of the $1S_5$ is drained off via the pumping process and the branching of spontaneous-emission decays.

A computer program has been written for an IBM 7090 calculator consisting of fifteen linear differential equations of the form

$$dN_i/dt = -N_i(C_i + \sum_j A_{ij}) + \sum_j A_{ji}N_j + \quad (10)$$

$$\sum_{j, i \neq j} B_{ij} U_{ij}(\nu_{ij})(N_j - N_i) :$$

the solution proceeds by the method of Laplace transforms. The populations of the ten $2p^5 3p$, the four $2p^5 3S$, and the ground state were considered. The A_{ij} are the spontaneous emission coefficients which may be obtained from

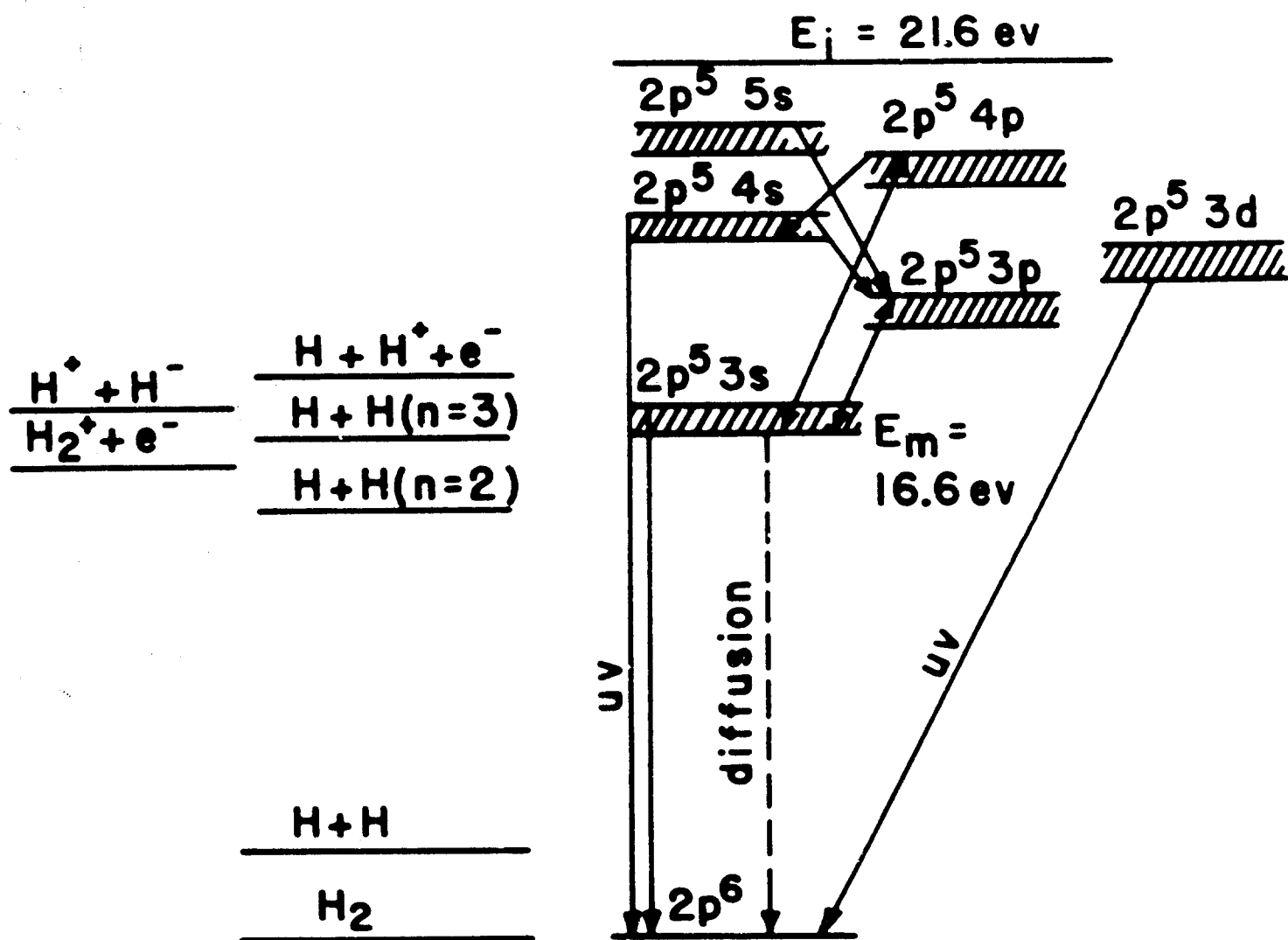
the measured oscillator strengths of the neon transitions.¹⁰ The B_{ij} are the induced emission/absorption constants and the $U_{ij}(\nu_{ij})$ is the radiant energy density at the transition frequency provided by the pumping lamp. The C_i are the non-radiative decay rates, which we may specify from independent experimental observations; they are, for example, $C(1S_5) = (350 \mu \text{ sec.})^{-1}$, $C(1S_4) = (110 \mu \text{ sec.})^{-1}$, $C(1S_3) = (280 \mu \text{ sec.})^{-1}$, and $C(1S_2) = (34 \mu \text{ sec.})^{-1}$, etc. The unknowns are the populations of these levels, given that at $t = 0$ when the flash is switched on, there is one particle in the $1S_5(2p^5 3S)$ state and zero in the rest. In Figures 7 and 8, we show results of the computer solutions for a square light pulse lasting $100 \mu \text{ sec.}$ corresponding to a radiation field of $\sim 5700^\circ \text{K}$. Although the problem has been considerably simplified, we believe the essential point has been made that a very modest illumination can appreciably alter the population and decay rate of the $1S_5$ state. The $2p_9(2p^5 3p)$ level, in particular, receives $\sim 2\%$ of the $1S_5$ population (Figure 9). Its density then becomes $\sim 10^9/\text{cc}$, while that of the $2p_4$ should be sufficient to switch off the lasing transition at 1.15μ or 0.633μ if the gas discharge tube be operated as a laser. The alterations of level population appear to occur irregularly in Figures 7, 8 and 9, owing to the superposition of the fifteen separate decay constants of the computer solution.

The above effects may be demonstrated with the red laser ($.6328 \mu$, $2p^5 5S \rightarrow 2p^5 3p(2p_4)$ transition as well in a short illuminated section of the classic He-Ne laser.

Upon being exposed to the Xe flash, it was observed that the light output of the laser was appreciably cross-modulated by the flash intensity (Figure 10). When the laser was oscillating vigorously and the flash intensity was comparatively weak, one observes that the cross-modulated decrease in the red laser's power is a faithful model of the flash intensity, demonstrating a response time of the order of a few microseconds. We believe that this quenching is caused by an increase of particles in the $2p_4(2p^5 3p)$ state, since the lifetime of this level is very short and consequently the laser power output (which depends on the population difference between the levels) follows the flash irregularities. However, when the flash illumination is intense and/or the oscillator is operating feebly, one finds a delayed and smoothed-over response on the cross-modulated laser output. We propose that this is caused by the quenching of the Helium 1S metastables by the radiation, with a corresponding decrease of particle density in the upper laser level. The 1S metastables can be pumped via the $^1S \rightarrow ^1P$ transition in helium, which occurs in a region of intense infrared output ($\sim 2\mu$) of the Xe flash. It is therefore reasonable to expect a response time $\sim 20 \mu$ sec. for the variations of the upper laser level, since this is populated by collisional activation with the long-lived He 1S state. Experiments with filtered flash radiation will hopefully establish these tentative conclusions in the near future, and observation of the side-radiated 6328A line from a He-Ne mixture will check the preceding assertion.

The success of the cross-modulation experiment suggests a potentially more useful technique with the infrared 1.15μ ($2p^5 4S \rightarrow 2p^5 3p$) laser transition. If the Xe flash radiation is filtered such that power at wavelengths only in the range $5000 - 3000 \text{ \AA}$ is available, then the $1S_5$ Ne metastable level will be pumped to the $2p^5 4p$ states preferentially with respect to the $2p^5 3p$ levels. A radiative side decay may then occur to the $2p^5 4S$ levels, one of which is the upper 1.15μ laser level. It is then clear that the presence of the Xe flash radiation under these circumstances causes the infrared laser to operate in a "closed-cycle" mode, with an improvement in output to be expected.

Experiments will also be conducted to ascertain the utility of these cross-modulation schemes for eventual commercial or military purposes. Since many gas lasers depend upon metastable excitation mechanism for operation, it is important to establish how the environment may affect the metastable concentration. We have shown that optical radiation may have an appreciable influence upon such a laser.



States of Hydrogen and Neon

Fig. 1

Neon 1.5 mm + 2.8 μ H₂

$n_e(t)$: decay of electron density in afterglow.

$N_m(t)$: decay of 3P_2 neon metastable density.

$H_\alpha(t)$: decay of $H_\alpha(6563\text{\AA})$ fluorescence.

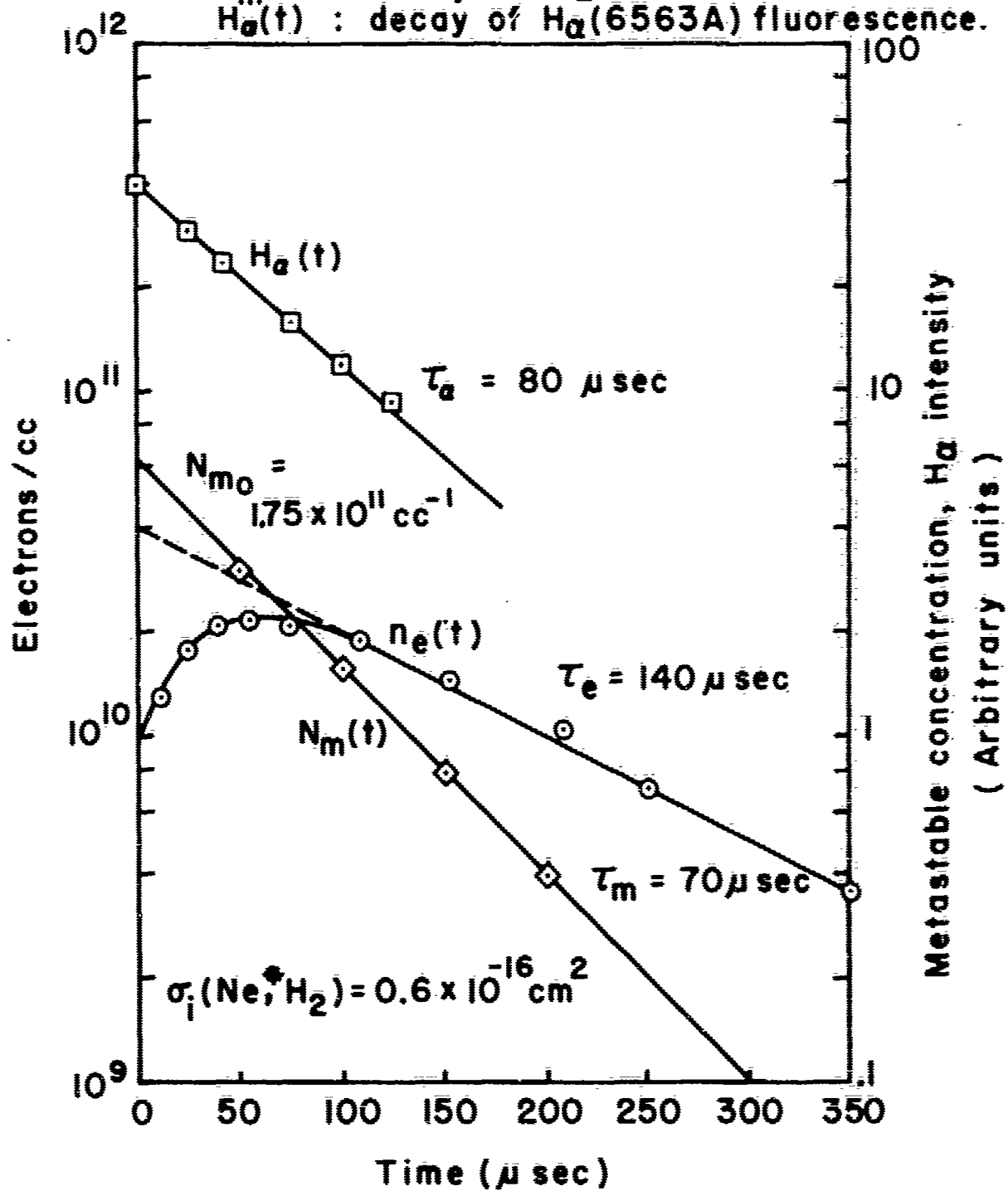


Fig. 2

DEACTIVATION RATE OF $\text{Ne}^*(3P_2)$ IN D_2
R.F. EXCITATION AT A CONSTANT LEVEL
NEON PRESSURE 1.5 mm

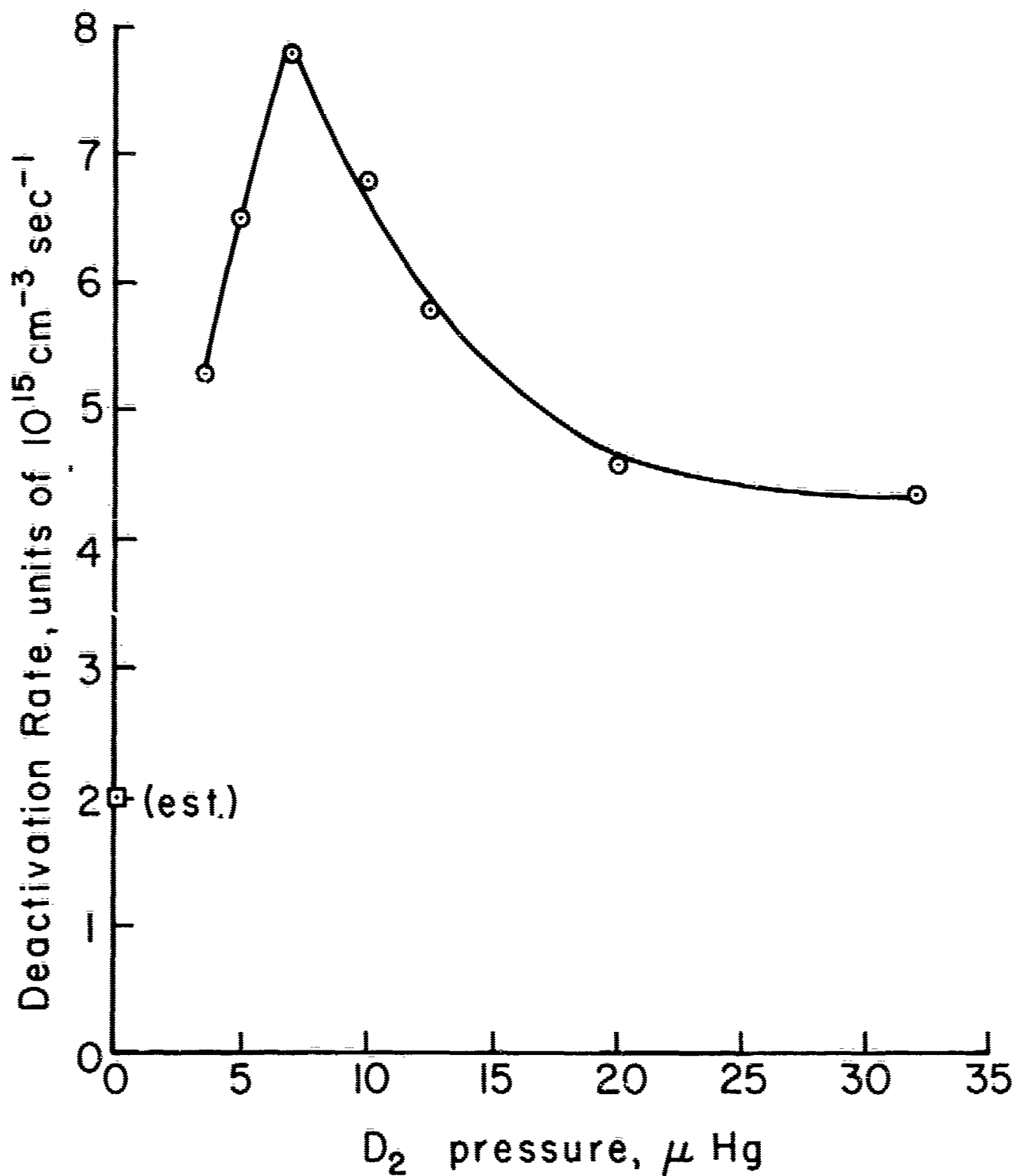
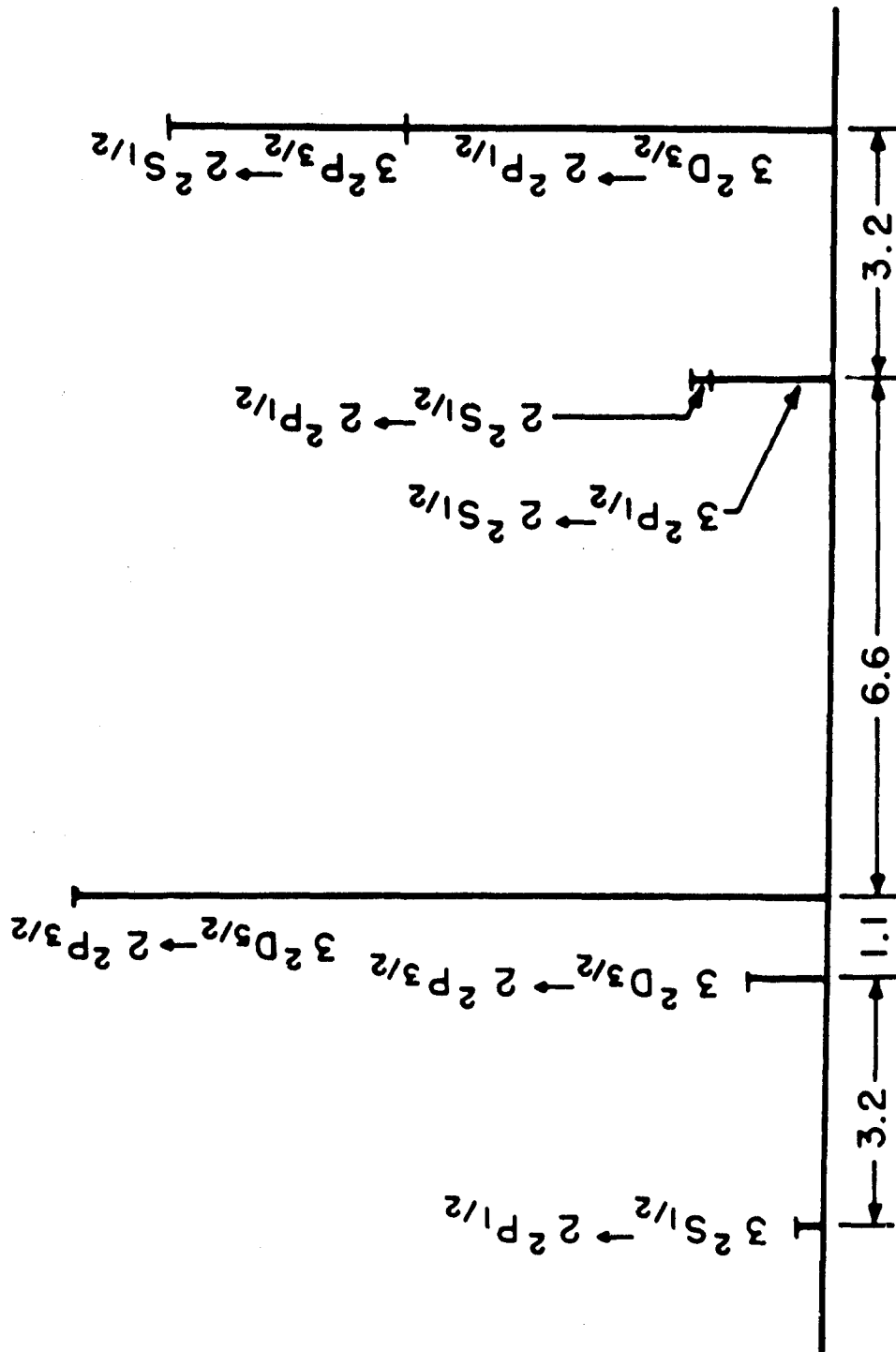


Fig. 3



FINE STRUCTURE OF $H\alpha$ (Separations in Gc)

Fig. 4

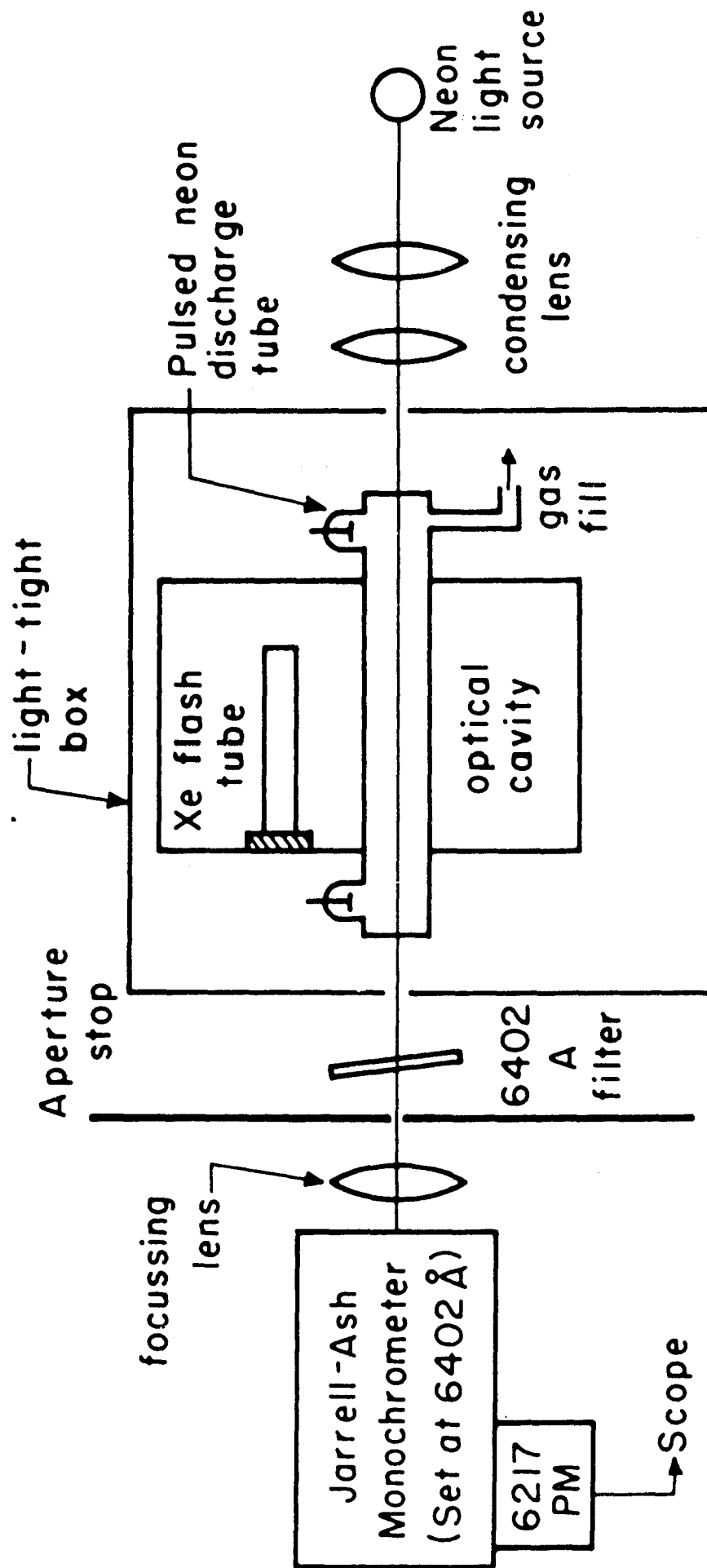


Fig. 5

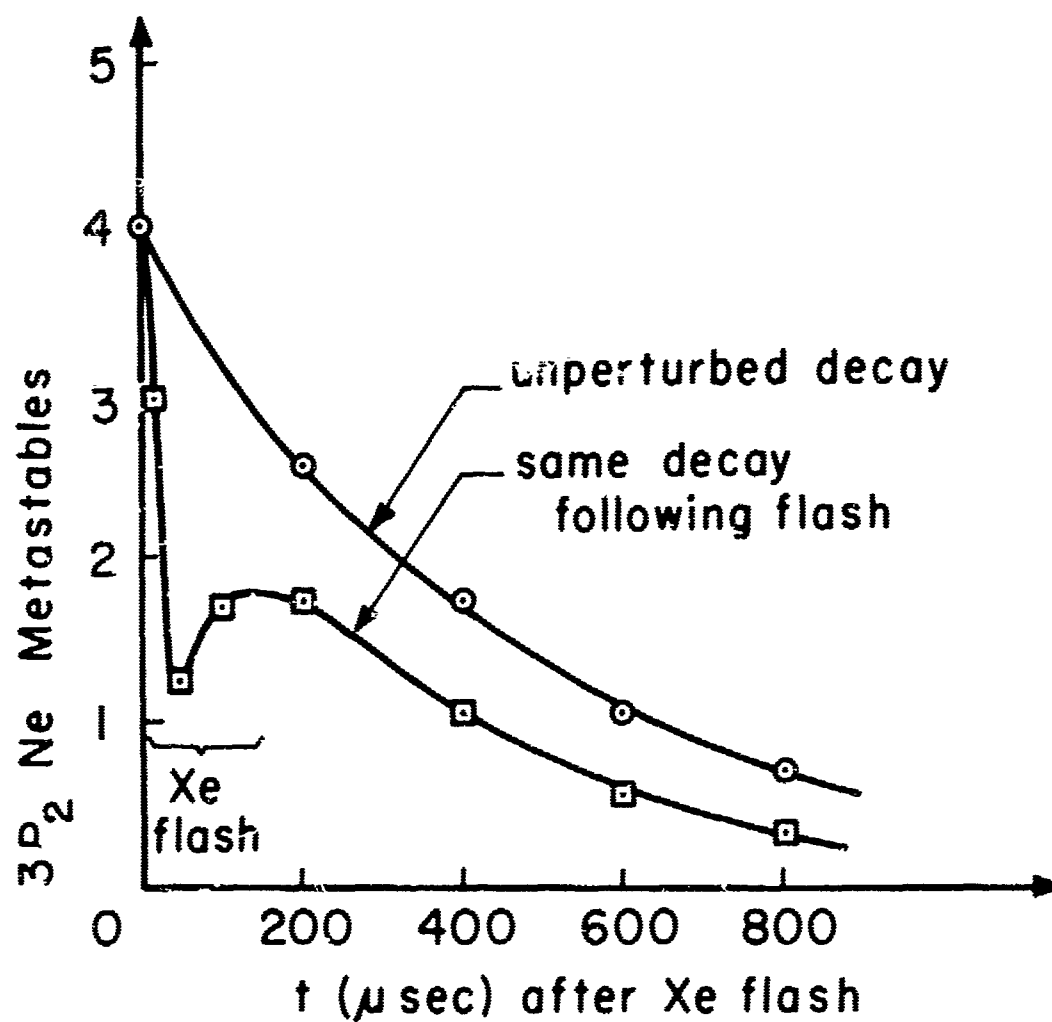
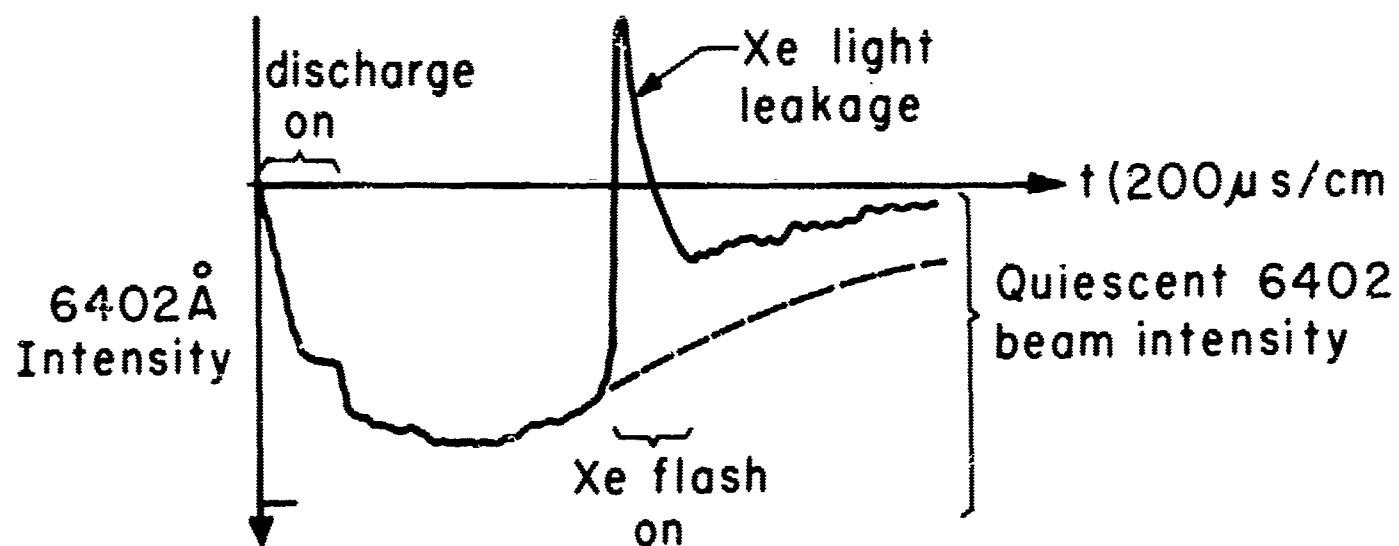
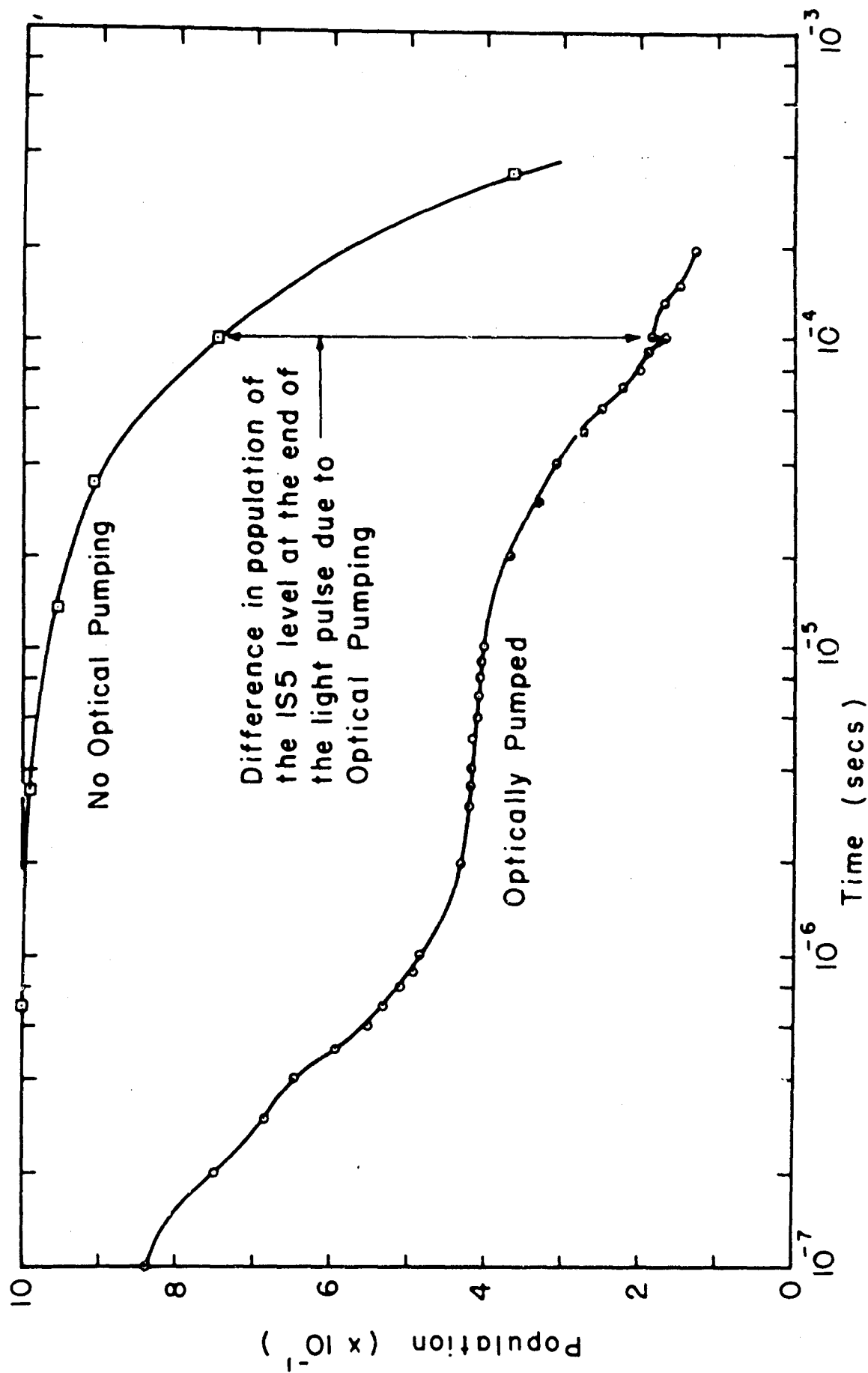
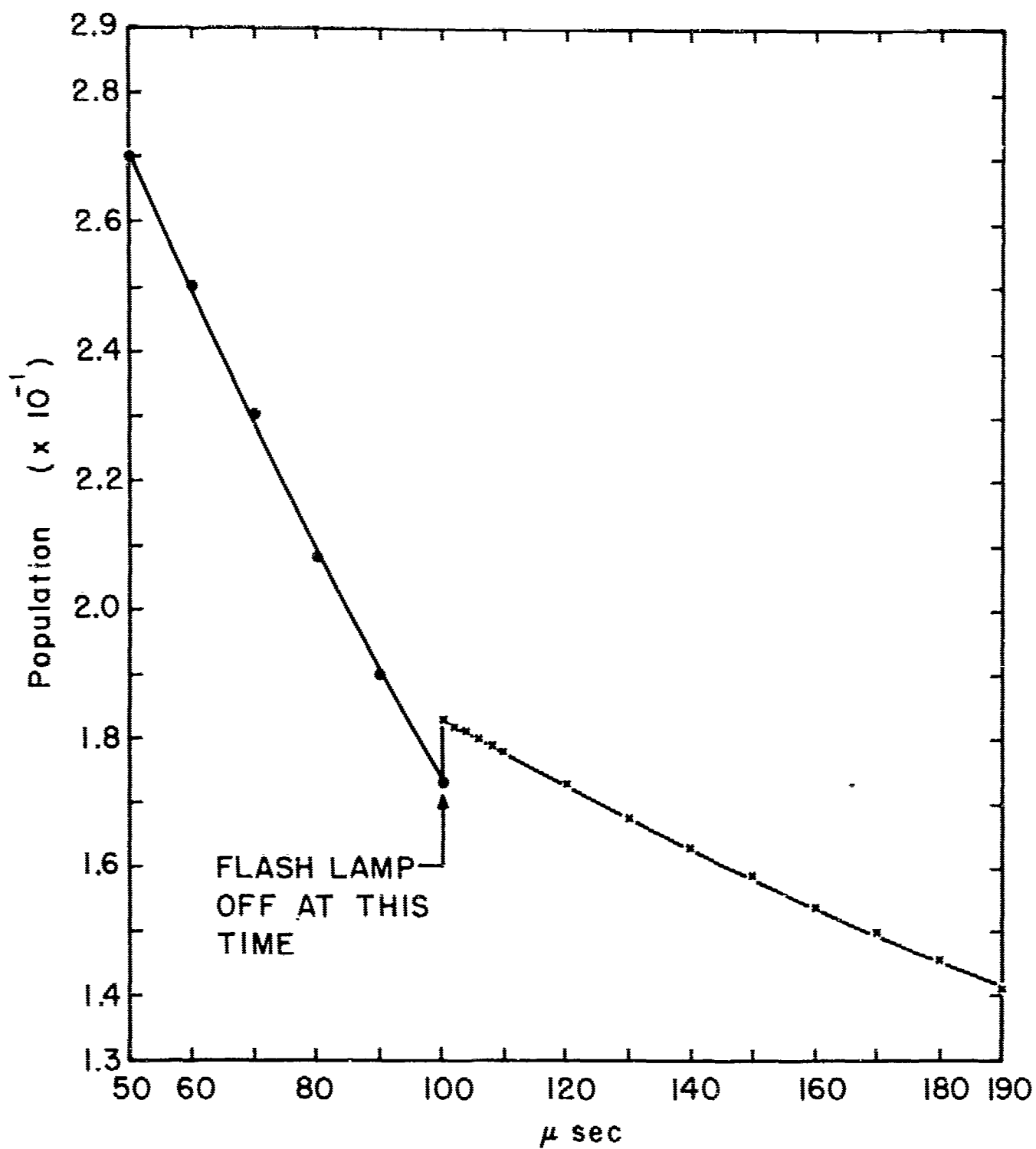


Fig. 6

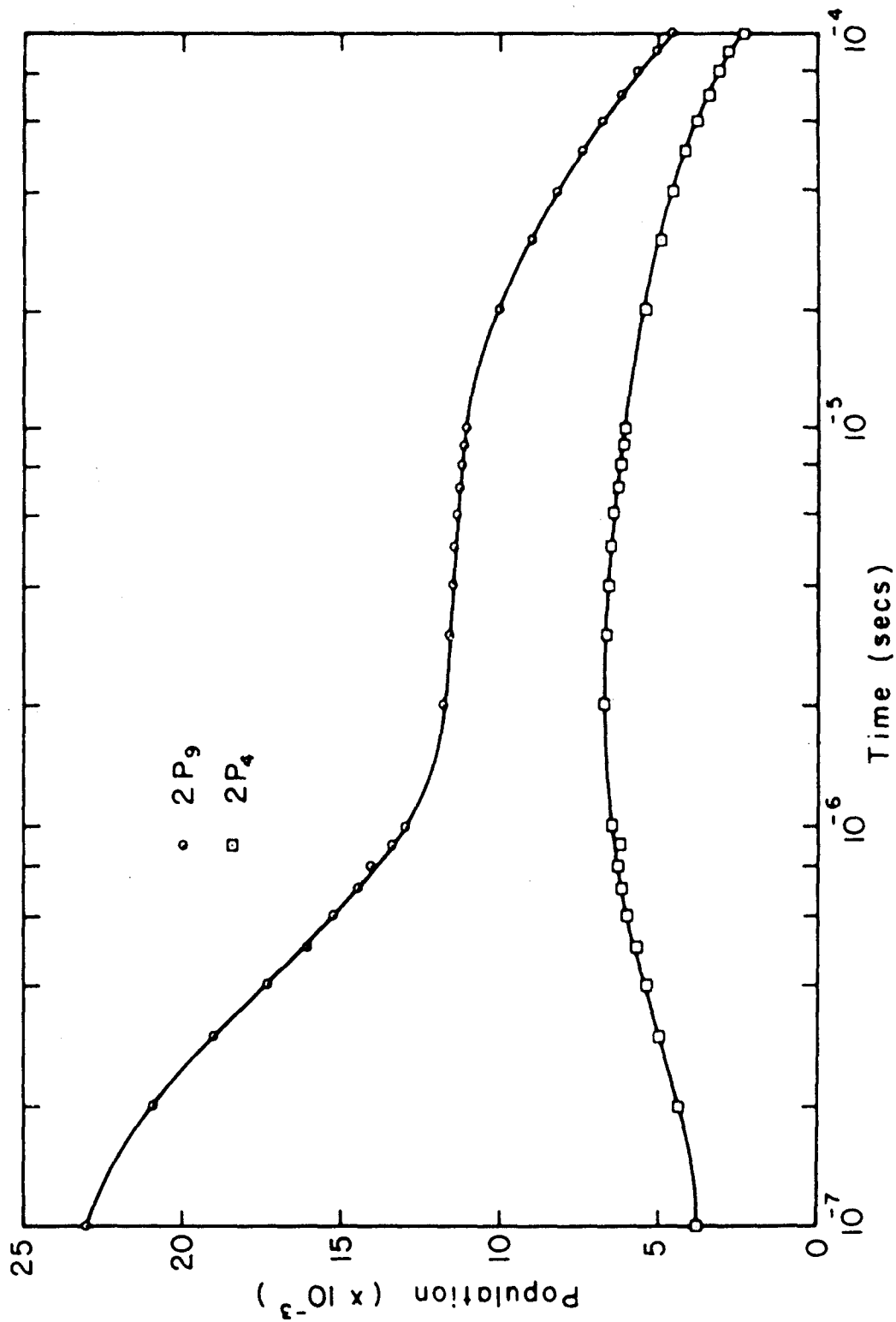


PREDICTED POPULATION OF THE 1S5 LEVEL
OF NEON WITH AND WITHOUT XENON FLASH.



PREDICTED POPULATION OF THE 1S5 LEVEL
OF NEON WITH XENON FLASH

Fig. 8



PREDICTED POPULATIONS OF LEVELS OF NEON DURING XENON FLASH.
 FLASH TEMPERATURE : 5700° K

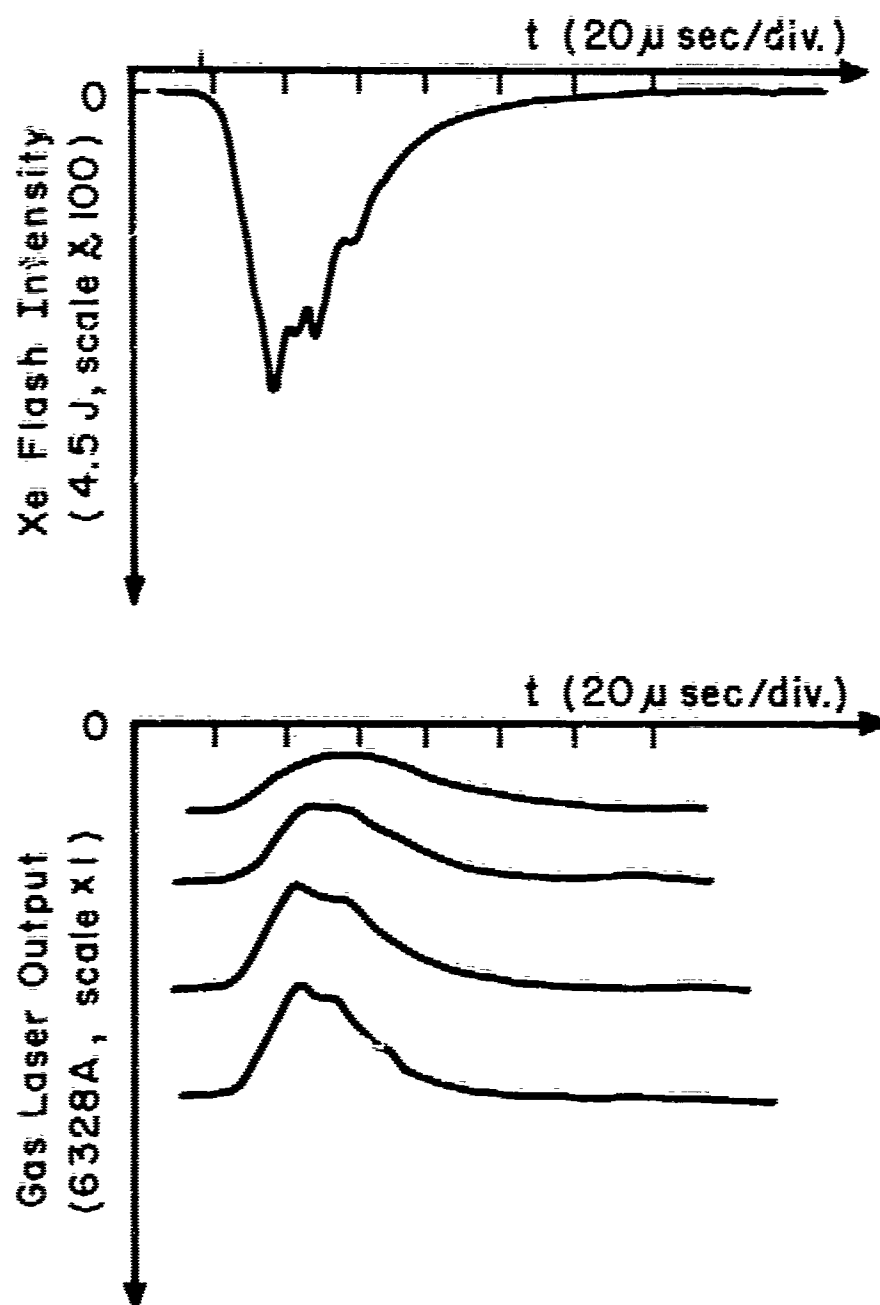


Fig. 10

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