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LASER PHYSICS: LIFETIME MEASUREMENTS, LINE PROFILES, HOLLOW CAT--ETC(U)
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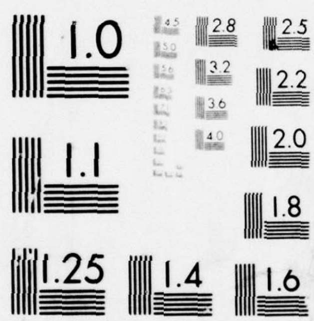

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Argon-Nitrogen Excitation Transfer
Mode-Locked Cavity-Dumped Lasers

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Research performed and published over the report period is summarized. Measurements of lifetimes in molecular Nitrogen laser levels, transfer cross sections in Ar-N2 mixtures, line broadening in Ne and Ar^+ are reported. Techniques for line profile analysis and gain measurement in exciplex laser systems are discussed. Properties of mode-locked cavity-dumped Argon ion laser pulses were measured and discussed.

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INTERIM REPORT AFOSR -74-2657C FOR THE PERIOD 15 JANUARY 1976 - 31 MAY 1977

A. RESEARCH RESULTS AND ACCOMPLISHMENTS

A paper describing precise measurements of pressure broadening on the $3s_2 \rightarrow 2p_n$ laser transitions of Neon was published (Ref.1). In this work, both Doppler and collision broadening was studied. Line-broadening parameters were extracted using the method of least squares in the computer analysis of spontaneous emission profiles transmitted through a scanning Fabry-Perot interferometer. ^{20}Ne-^{4}He and ^{20}Ne-^{20}Ne Lorentz broadening parameters were tabulated and their temperature dependence summarized. (Ref.1.)

A paper reporting a novel method for the design of mode-locked cavity-dumped lasers was completed and published. Our approach makes use of generalized confocal-equivalent mode theory to design folded cavities with harmonically related repetition frequencies so that acoustic modulators and deflectors with differing fundamental resonances can be incorporated in the same mode-locked cavity-dumped laser. (Ref.2.)

A detailed series of measurements of second-order autocorrelation functions for single pulses extracted from mode-locked cavity-dumped lasers is nearly completed and a paper describing this work is in preparation. The method of measurement makes use of the two-quantum photoeffect and was developed during our previous AFOSR grant at Yale [see, W.R.Bennett,Jr. et al, IEEE J.Quant. Elect. QE-10, 97 (1974)]. OUr present measurements have been made as a function of cavity loss, laser amplifier gain and line width. Careful allowance for effects of lineshape have been included in the comparison of experimental pulse widths with theoretical results. It is our conclusion that the mode-locked cavity-dumped laser pulse shapes are bandwidth limited to gain profiles in close agreement with our previous studies of Argon ion laser transition lineshapes [see, R.C.Sze and W.R.Bennett,Jr., Phys.Rev. A, 5, 837 (1972)]. These comparisons were effected in the following manner: a) The previous line broadening and gain profile

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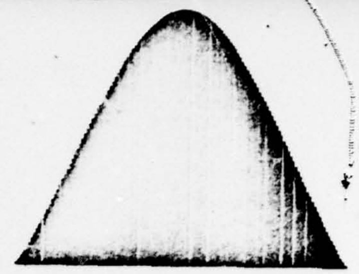
data were used to determine the number of simultaneously oscillating modes that should have been excited for a given cavity loss as a function of discharge current. The power distribution in this mode distribution spectrum was then compared with a direct analysis of the mode-locked laser distribution determined with a scanning Fabry-Perot interferometer. b) The computed mode distribution was then used to determine the shape of the mode-locked laser pulse that should result from the previous gain-bandwidth data by taking the Fourier transform of the mode-locked laser mode distribution. (See Fig.1.) Finally, the full width at half maximum of the computed intensity pulse was compared with the same quantity for a pulse which was deconvolved from the measured second-order auto-correlation intensity function. (See Fig.2.) A detailed discussion of these results will be included in D.B.Carlin's PhD dissertation and will be submitted for publication shortly. (Ref.3) We are currently pursuing an experiment in which these pulses (which are significantly shorter than had been suspected by most previous investigators) will be used to probe the lifetime of the lower states of the argon ion laser transitions. (These lifetimes are in the subnanosecond range and would be difficult to determine by conventional electronic detection methods.)

A direct determination of the lower state lifetimes in the argon ion system is important for several reasons. These lifetimes are key parameters in understanding this important laser system, and there has been an unfortunate tendency of both theorists and experimentalists to report contradictory results for the quantities in question. The initial calculated values by Statz et al [J.Appl. Phys. 36, 2278 (1965)] were in error by a factor of five due to an error in statistical weight assigned to the Ar^+ ground state configuration [see, Statz et al, J.Appl.Phys. 39, 4045 (1968)]. More recently, Van der Sijde et al [J.Quant. Radiat.Transfer 16, 1011 (1976)] have reopened the controversy by reporting Fabry-Perot measurements of Lorentz widths on the argon ion transitions which are substantially in excess of the low current values reported earlier by Sze and Bennett [Phys.Rev.A, 5, 837 (1972)]. (Van der Sijde et al were evidently not

a) Mode Distribution
(Computed from measured linewidths and cavity loss).



b) Computed Laser Pulse
(Assuming mode distribution shown above)

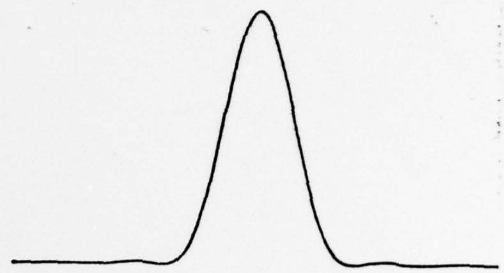
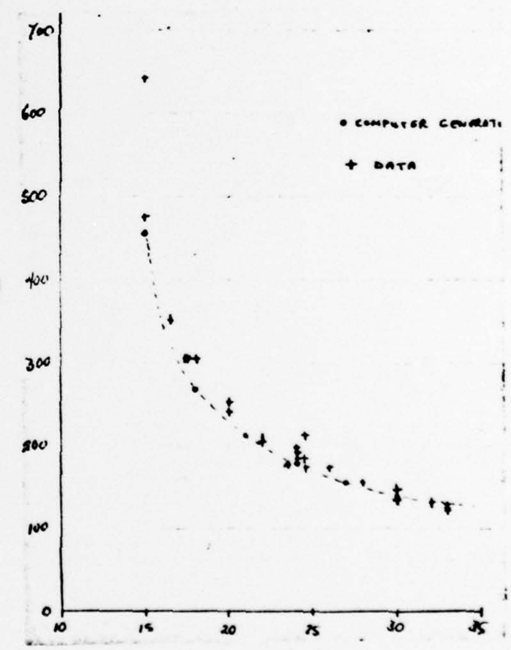


Fig.1. a) Mode intensity distribution for a Voigt profile using measured linewidth data and known cavity loss.

b) Intensity pulse as function of time, computed from Fourier transform of phase-locked mode distribution shown in a).

Full Width at Half Maximum (in picosecs) for Intensity Pulse



Discharge Current
(in Amperes)

Fig.2. Comparison of measured [by deconvolving the second order auto-correlation function, $G^2(\tau)$] and computed pulse widths for a mode-locked cavity dumped argon ion laser oscillating at 5146 Å vs. discharge current. The computed results were based on data of the type shown in Fig.1. (+ = measured; • = computed).

even aware of the earlier measurements by Sze and Bennett.) Because the method of Fabry-Perot analysis we developed earlier with AFOSR support has the potential of becoming an accurate plasma diagnostic method through linebreadth studies, it seemed important to clarify questions that might be raised regarding the accuracy of our method. In anticipation of this type of question we have indeed made direct determinations of Lorentz widths on the 4880 Å transition in the argon ion laser using a spectral hole-burning technique. (A strong running wave is used to burn a hole in the gain profile and a weak running wave travelling in the opposite direction is used to measurement the decrease in gain across the hole; the technique was originally developed under AFOSR support at Yale and is similar to the "saturated absorption spectroscopy" technique used very successfully by Hänsch and others.) We are currently in the process of writing up these results for publication. We get reasonably good agreement with our previous values for the 4880 Å Lorentz widths in the medium-to-high current regime at low pressures. The values typically are ≈ 850 MHz at currents in excess of 15 A/cm^2 and filling pressures ≈ 0.3 Torr. These numbers agree reasonably well with those of Van der Sijde et al, which were evidently taken at lower pressure and higher current. The values fall in the plateau region previously reported by Sze and Bennett. However, a marked decrease in Lorentz widths was found with the hole-burning technique for currents below ≈ 10 Amperes/cm^2. Unfortunately, the sensitivity of the hole-burning method also falls off drastically in the same current range and accurate zero-current intercepts were not obtainable with the hole-burning method. Hence, all three sets of measurements agree reasonably well at moderate to high currents. But to date, the low-current intercepts have only been observable with the enormously more sensitive computer oriented method used by Sze and Bennett. (At low currents, the excited state densities are varying with the square of the current; hence, a slight decrease in current produces a huge decrease in experimental signal.) (Ref.4.)

A summary of research on cavity modes and hole burning effects supported by AFOSR at Yale is to be published in July 1977 as a book entitled The Physics of Gas Lasers by W.R.Bennett,Jr. (Gordon and Breach, Science Publishers, London). This work represents a revised and updated edition of a previous paper entitled "Some Aspects of the Physics of Gas Lasers" that had been included in the Brandeis University series on Atomic Physics and Astrophysics. The new volume will be issued as a separate technical report to AFOSR when it arrives from the publisher. (Ref.5.)

We completed a series of measurements of lifetimes of N_2 and N_2^+ molecular states which are important for laser action. These measurements used pulsed excitation by threshold energy electrons and multi-channel delayed coincidence photon counting. New time-interval measurement circuitry was incorporated in the experiment over that which had originially been developed by Bennett and Kindlmann. [see, e.g., Phys.Rev. 149, 38 (1966).] The present system incorporates an on-line computer-controlled 500 MHz counter as the primary time interval measuring device and computer-controlled pulse generators and data processing equipment. The present apparatus represents a substantial improvement in sensitivity and flexibility over the older vernier chronotron method that we used for this sort of measurement in the past. Threshold for direct excitation to the desired state is determined by measuring the counting rate as a function of pulse height and data are taken close to threshold (typically within 0.2 eV). The data are then least-squares fit to appropriate theoretical forms using the same computer arrangement. Decay rates determined in this manner are plotted as a function of pressure to determine both the radiative decay rate (zero-pressure intercept) and collision deactivation cross section (from the slope of the pressure plot). The values for N_2 radiative lifetimes determined in this way are shown in Table I. Substantial differences were encountered between our measurements and previously reported values. We attribute these differences to radiative cascade effects in the earlier work. (Ref.6.)

Table I. Radiative lifetimes and limits of error determined for important levels in the N_2 and N_2^+ laser.

State	Lifetime (nanoseconds)
$C\ ^3\Pi_u (v'=0)$	37.6 ± 1
$C\ ^3\Pi_u (v'=1)$	40 ± 3
$B\ ^2\Sigma_u^+ (v'=0)$	37.9 ± 1.8

Most of our recent work has been devoted to the study of electron transfer rates from noble gas metastable states to a second gas component. In this work selective excitation of the initial (metastable) level (level 1 in the following notation) is provided by threshold energy electron impact in short excitation pulses. The collision transfer rate per atom (a_{12}) is measured through radiative decay (at rate A_2) of the second component excited state using our delayed coincidence photon counting methods. The main features of the process are included in the general two-level model shown in Fig.3.

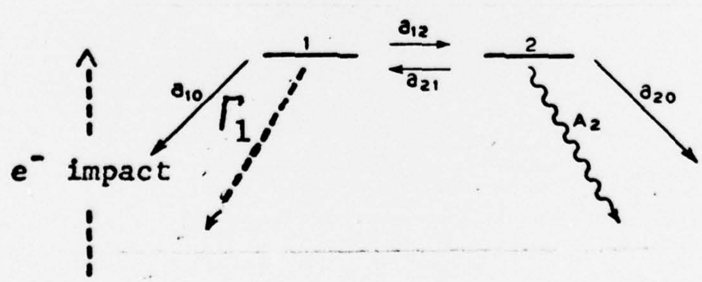


Fig.3. General two-level collision transfer model used to analyze the present experiments. The metastable level (1) is excited by threshold energy electron impact. The process is detected by the total decay rate from level 2 using radiative decay at rate A_2. Γ_1 is the diffusion rate of the metastable and a_1, a_2 are collision destruction rates for levels 1 and 2.

The level densities n_1 and n_2 are coupled through the equations

$$\dot{n}_1 = -(\Gamma_1 + a_1)\, n_1 + a_{21} n_2$$
$$\dot{n}_2 = -(A_2 + a_2)\, n_2 + a_{12} n_1 \tag{1}$$

after the short pulse of electron excitation is turned off. Here, the quantities are defined as stated in the caption to Fig.3, with the addition that a_1 and a_2 represent the sumsof all two-body collision destruction channels including the primary transfer rates of interest (a_{12} and its inverse, a_{21}). The solutions for n_1 and n_2 are given by the sum of two exponential terms with decay rates

$$R_1 = \frac{\Gamma_1+A_2+a_1+a_2}{2} + \frac{\Gamma_1-A_2}{2}\sqrt{1 + \frac{2(a_1-a_2)}{(\Gamma_1-A_2)} + \frac{(a_1-a_2)^2+4a_{12}a_{21}}{(\Gamma_1 - A_2)^2}}$$

and

$$R_2 = \frac{A_2+\Gamma_1+a_2+a_1}{2} + \frac{A_2-\Gamma_1}{2}\sqrt{1 + \frac{2(a_2-a_1)}{(A_2-\Gamma_1)} + \frac{(a_2-a_1)^2+4a_{21}a_{12}}{(A_2 - \Gamma_1)^2}} \tag{2}$$

Expanding the exact solutions in Eqs.(2) through second order terms in the pressure,

$$R_1 = \Gamma_1 + a_1 - \frac{3(a_1-a_2)^2+16a_{12}a_{21}}{4(A_2 - \Gamma_1)} + \text{Order } (a^3) \quad \text{(Slow)}$$

and

$$R_2 = A_2 + a_2 + \frac{3(a_1-a_2)^2+16a_{12}a_{21}}{4(A_2 - \Gamma_1)} + \text{Order } (a^3) \quad \text{(Fast)} \tag{3}$$

Thus the decay of the radiating second component is described by the sum of a slow and a fast exponential component, and so long as A_2 is not at all comparable to Γ_1, a linear pressure dependence of the slow decay rate at low pressures occurs which permits a precise determination of the collision transfer cross section. The coefficients $a_1=a_{10}+ a_{12}$ involve the appropriate ground state densities, relative velocities and velocity averaged cross sections. Specifically,

$$a_{12} = 0.81 \times 10^6\, P_2\, \sigma_{12}\sqrt{\left(\frac{300}{T}\right)\left(\frac{M_1+M_2}{M_1 M_2}\right)} \quad \text{sec}^{-1} \tag{4}$$

where P_2 is the partial pressure of the second component in Torr. σ_{12} is the cross section for rate a_{12} expressed in units of 10^{-16} cm^2,

M_1 and M_2 are the masses of the two gas components expressed in amu, and T is the absolute temperature. In practice "Level 2" must be regarded as any group of electronic levels of the second gas component which is in close (say ≈ 1 eV) energetic coincidence for energy transfer from Level 1. Under these circumstances σ_{12} represents a total transfer cross section to the group of levels involved. However, it then becomes especially important to take data at low enough pressures to insure the absence of quadratic pressure dependent terms in the excited state decay rates. (Most data previously reported in the literature of two-component gas laser systems have been extracted in a manner which totally ignores these fundamental requirements on pressure, not to mention equally important requirements on selective excitation of the initial state.) Except in the case of Penning ionization, the destructive collision rate through non-resonant channels (i.e., the rate a_{10}) tends to be negligible at low pressures. For example, with the $Ar\text{-}N_2$ data described below, reactions of the type

$$\begin{aligned} Ar^* + N_2 &\rightarrow Ar + N_2 + 11.5 \text{ eV} \quad \text{(Kinetic Energy)} \\ Ar^* + Ar &\rightarrow Ar + Ar + h\nu \\ Ar^* + 2Ar &\rightarrow Ar_2^* + Ar \end{aligned} \qquad \text{(A)}$$

are quite negligible compared to the reaction

$$Ar^* + N_2 \rightarrow N_2^* \, (C^3\Pi_u) + Ar \qquad \text{(B)}$$

of primary interest. Representative data for reaction (B) taken with our apparatus are shown in Figs. 4 and 5. These figures show the time-dependent decay of the $C^3\Pi_u(v'=0)$ level of N_2 when excited by $Ar(^3P_2)$ metastables in $Ar\text{-}N_2$ mixtures at two different pressures. Both the signal and log of the signal are shown plotted against time. (The feducial marks on the vertical axis represent 1/e points for the Log plot.) As is readily seen from the data, the level decay is clearly characterized by two widely different decay rates after the electron pulse is turned off. The slow component decay rates were extracted using a least squares fit to a functional form consisting of the sum of two exponential decaying terms plus a background constant.

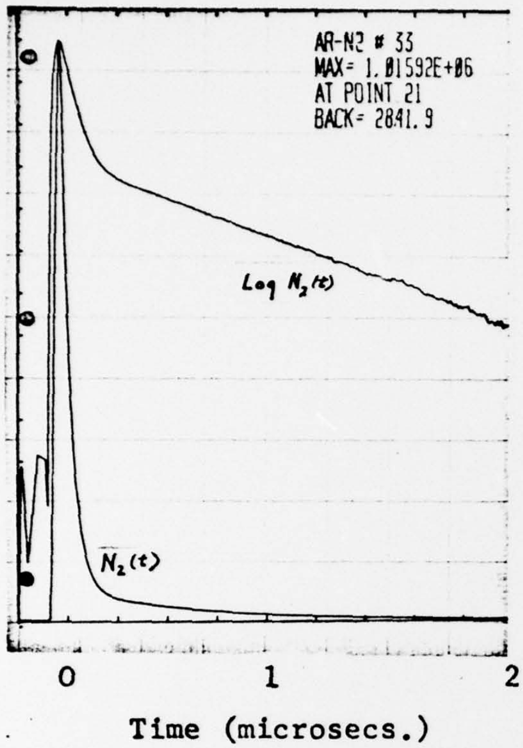


Fig.4. N_2 $C^3\Pi_u$ ($v'=0$) as function of time (from λ3371 Å) after threshold energy electron pulse for Ar($^3P_2$) metastable is turned off in Ar - N_2 mixture. (Ar at 1.66 Torr and N_2 at 1.8 Torr at 663 °K.)

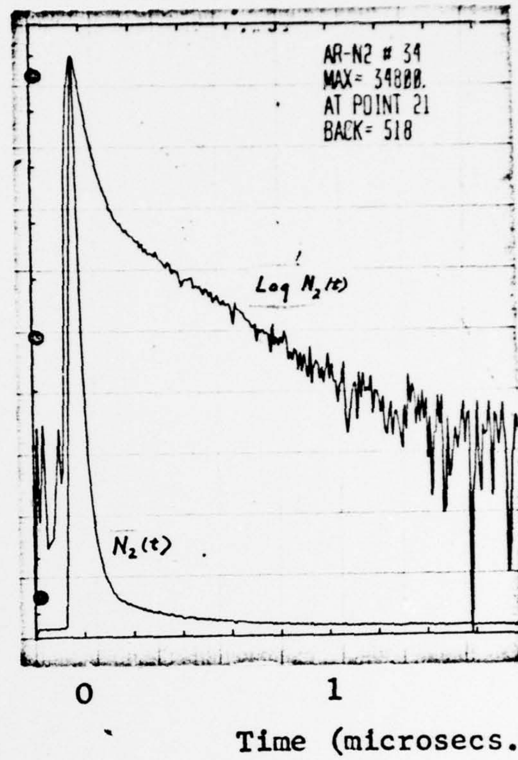
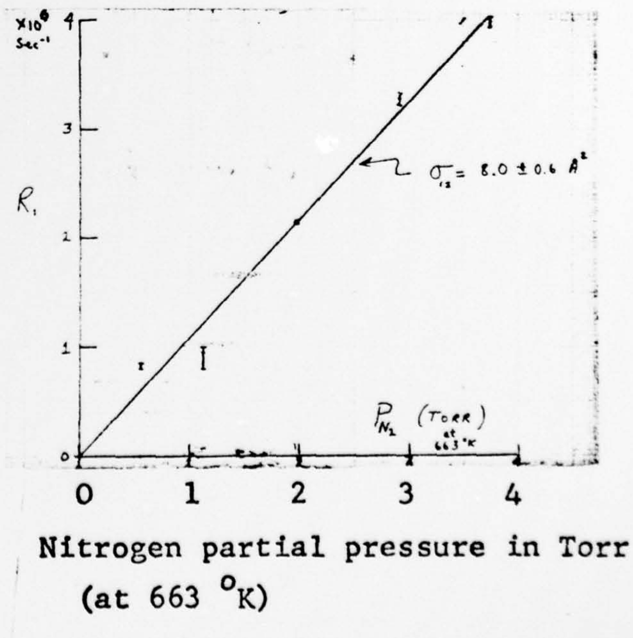


Fig.5. Same as Fig.4, except N_2 pressure was 2.65 Torr

Fig.6. Variation of the decay rate of the slow exponential component extracted from data such as that in Figs. 4 and 5, shown as a function of nitrogen partial pressure. The straight line corresponds to an excitation transfer cross section from the Ar metastables of 8.0 Å^2. (Error bars are standard deviations in least-squares fit.)

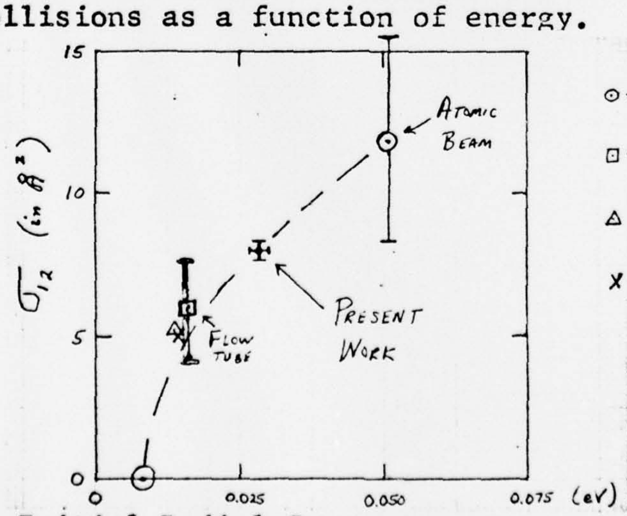


Values for the decay rates of the slow component are shown in Fig.6 as a function of nitrogen partial pressure, for constant argon pressure (1.66 Torr) and temperature (663 ^{o}K). From Eq.(4), we then extract values of the excitation transfer cross section from the argon metastables. The results so far imply a cross section of 8.0 $\pm$ 0.6 Å ^{2} for reaction (B). We believe that this is the most accurately determined value for this cross section to date. The different measurements imply a large variation of the cross section with mean relative initial energy, hence gas temperature. The latter is quite surprising for an exothermic reaction and may just be the result of error. A comparison of several measurements is given below in Fig. 7. The results for the cross section are expressed in Å^2 and shown plotted as a function of the mean relative energy of the radial component of the motion as seen initially in the center of mass system. This relative kinetic energy is simply kT/2 when both gas components have Maxwellian velocity distributions at the same temperature. This method of expressing the data was needed to permit comparison with the cross section obtained from a crossed atomic beam experiment. We are currently writing these results up in more detail for publication. After completing the Ar-N_2 work we want to take similar data on charge transfer reactions important to the N_2^+ laser. For example, there is a complete lack of data on the reaction

$$Ne^+ + N_2 \rightarrow N_2^{+\,*}(B\ {}^2\Sigma_u^+) + Ne$$

which is potentially of importance to the laser transition at 3914 Å.

Fig.7. Excitation transfer cross-section for Ar($^3P_2$)-N_2 collisions as a function of energy.



⊙ - Winicor and Fraites, J. Chem. Phys. 61, 1548 (1974).

□ - Piper et al, J.Chem.Phys. 59, 3323 (1973)

△ Calo and Axtmann, J.Chem.Phys. 54, 4961 (1971).

X Bourene and LeCalvé, J.Chem. Phys. 58, 1446 (1973).

Initial Radial Component of Mean Relative Energy.

We have developed two different methods for the measurement of gain in exciplex laser systems. Because many potentially interesting exciplex systems involve corrosive gases, it seemed desirable to determine their gain characteristics first in disposable glass or quartz cells before attempting to use such systems in oscillating lasers. Both methods are capable of determining gain coefficients with a long-term rep ducibility ≈ ± 0.1 percent per pass. Both methods incorporate laser sources which at the moment can only be tuned in discrete jumps over oscillating lines characteristic of the source. However, either method could easily be converted to more widely tunable dye laser sources should the need arise. Many exciplex systems have broad enough emission bands to afford coincidences with discrete lines in the argon ion and N_2 laser, and we are investigating these exciplex systems first. Method 1) makes use of repetitive, short pulses from a TEA laser to probe cw or quasi-cw gain in a test cell. Method 2) uses a cw ion laser (tunable over a range of discrete lines by means of an internal prism) to probe a pulsed, or otherwise modulated, test cell. The two methods are outlined schematically in Figs.8 and 9. In each case the same detector is used to measure the original source laser intensity and the amplified source laser by time modulation techniques. In method 1) the reference pulse is run through an optical delay in excess of the pulse duration before entering the photodetector. Two pulses hit the same detector in a time-resolved fashion and are then fed to separate A-to-D converters for computer averaging. In method 2), the gain tube is modulated and a comparison of the detector signal is made at different times through use of a multichannel waveform integrating circuit.

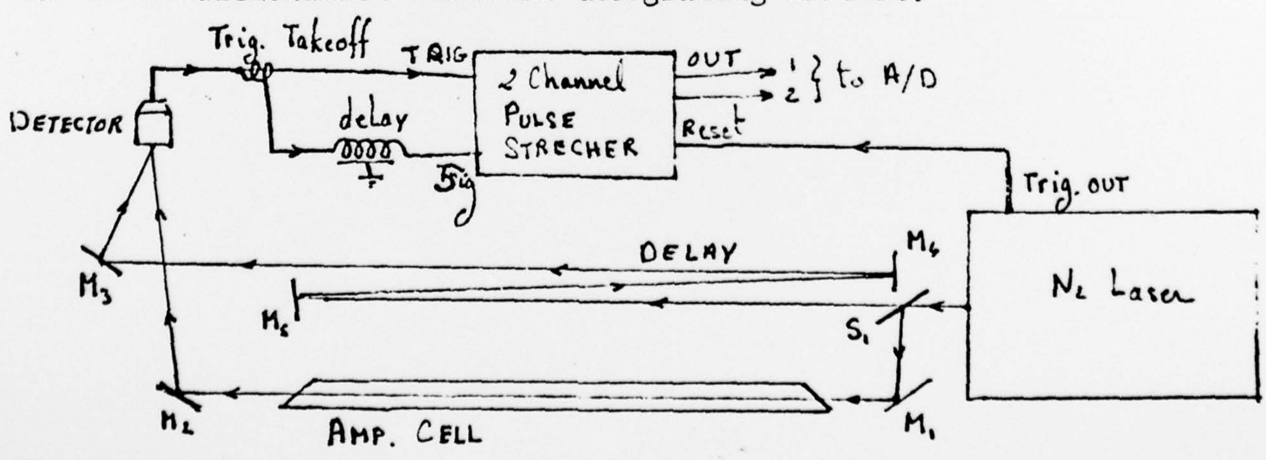


Fig.8. Schematic diagram of Method 1). A pulsed laser source is used to measure gain in a cw or quasi cw test cell.

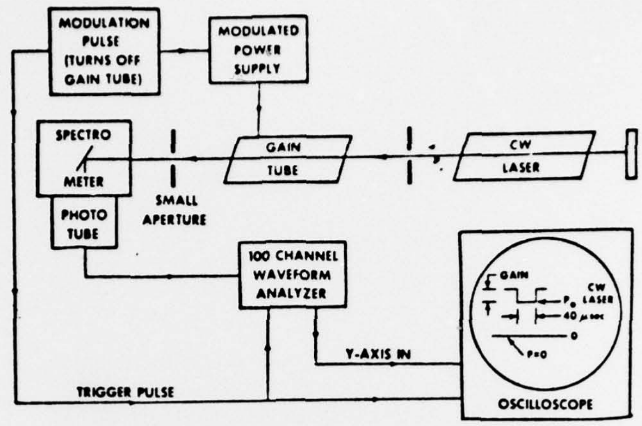


Fig.9. Schematic diagram of Method 2). A cw tunable laser is used to measure gain in a pulsed or modulated test cell.

We are currently using Method 1) to probe gain and absorption in the Ar-N_2 system and are using Method 2) to pursue gain measurements in various exciplex systems of the type discussed in renewal proposal for the present research contract. We have been joined in this activity by Prof. Santaram Chilikuri (currently on leave from Union College), who has done previous spontaneous emission spectroscopy on various exciplex emission bands that might make suitable laser systems.

A Fourier transform method for line profile analysis has been studied and an article describing the technique is to be published. The suitability of the method as a passive probe to determine laser plasma characteristics is currently under investigation.

B. PUBLICATIONS

1. J.W.Knutson,Jr. and W.R.Bennett,Jr., "Line Broadening of the $3s_2$-$2p_n$ Laser Transitions of Neon", Phys.Rev.A 13, 318 (1976).

2. D.B.Carlin and W.R.Bennett,Jr., "Mode-locked, Cavity-dumped Laser Design Considerations", Appl.Opt. 15, 2020 (1976).

3. D.B.Carlin and W.R.Bennett,Jr., "Characteristics of Mode-Locked Cavity-Dumped Lasers" (to be published).

4. B.C.Wexler and W.R.Bennett,Jr., "Hole-burning Method for the Analysis of Line Widths" (to be published).

5. W.R.Bennett,Jr., The Physics of Gas Lasers (Gordon and Breach, London. Currently scheduled to appear in July, 1977).

6. J.Flint and W.R.Bennett,Jr., "Radiative Lifetimes of Molecular Nitrogen Laser Levels". (to be published)

7. W.R.Bennett,Jr.,and J.Flint, "Excitation Transfer Cross Section in Argon-Nitrogen Mixtures". (in preparation).

8. W.R.Bennett,Jr., "Fourier Analysis of Line Profiles". (to be published).

C. PEOPLE PARTICIPATING IN RESEARCH CURRENTLY

Dr. W.R.Bennett,Jr., Principal Investigator, C.B.Sawyer Professor of Engineering and Applied Science and Professor of Physics.

Dr. Santarm Chilukkuri, Associate Professor of Physics on leave from Union College, currently,Visiting Professor of Engineering and Applied Science at Yale.

Mr.D.B.Carlin, graduate student in Engineering and Applied Science, PhD expected in Sept., 1977.

Mr.S.Jabr, graduate student in Engineering and Applied Science.

Mr.J.Flint, graduate student in Engineering and Applied Science.

Mr.R.Fortier, undergraduate research assistant in Engineering and Applied Science.

W.R. Bennett, Jr.