SEMI-ANNUAL TECHNICAL STATUS REPORT

ON

MOLECULAR LASER STUDY

3-5 MICRONS

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1.0 INTRODUCTION

This first Semi-Annual Technical Report is in partial fulfillment of the contract requirements as specified in the Contract Schedule, Section G, Paragraph I(B).

Since this is the first technical report written on this contract, it will also present some pertinent background on molecular lasers. This will be in the form of a brief review of N_2-CO_2-He lasers and the operation of our plasma tubes to be presented in Section 2.0. Discussion of laser emission from a mixture of H_2-C_2H_2-He will be presented in Secti

Section 4.0 will outline the proposed experimental w to be carried out in the second half of the contract period.

The operation of the first acetylene laser and identification of the lasing transition were carried out by Dr. Carl F. Shelton. He initiated the experimental program at IBM and was the Principal Investigator for the first quarter of the performance period for this contract.

2.0 BRIEF REVIEW OF N_2-CO_2-He LASERS

The pumping mechanisms and operation of the N_2-CO_2-He laser system have been widely discussed in the open literature and thus will only be summarized briefly here.

Excitation mechanisms for pumping CO_2 to the upper laser level include:

(a) Direct electron impact[1],

(b) Vibrational energy transfer[2] through inelastic collisions with $N_2^*(v = 1)$,

(c) Recombinations[3] of the form

$$CO + O \rightarrow CO_2^*(001)$$

Helium enhances the CO_2 laser output by at least five mechanisms:

(1) Helium depopulates the 01^10 mode through inelastic collisions and thus indirectly increases the rate of depopulation of the lower laser level, i.e., the $10^0$0 level[4].

(2) Helium increases the rate of rotational thermalization within each vibrational level and thus maintains the population inversion on the strongest vibrational-rotational transitions.

(3) Helium cools the kinetic temperature of the gas mixture because of its higher thermal conductivity.

(4) Helium shifts the electron temperature and/or the electron density to more favorable values for vibrational excitation by electron impact[4, 5].

(5) Helium reduces the diffusion of the excited species to the walls of the plasma tube where they can be de-excited.

Effect (2) shows up in the fact that fewer rotational P-branch lines are observed in N_2-CO_2-He mixtures than in N_2-CO_2 mixtures. Patel[3] observed emission on the P(12) through P(38) lines of the $\nu_3 - \nu_1$ band with pure CO_2. Other researchers have observed emission on the P(18) through P(28) lines using

N_2-CO_2[6], the P(20) through P(26) lines using CO_2-He[7] and the P(20) through P(24) using N_2-CO_2-He[8]. Again, since only the even P-branch lines are allowed this means a reduction from fourteen lines using pure CO_2 to four lines using CO_2-He and to three lines using N_2-CO_2-He.

The actual effects of He on electron temperature and electron density in the plasma have not been accurately measured during laser emission, however, these effects can be inferred in various ways. Patel[9], for example, observed an increase in output from his parallel pumped N_2-CO_2 laser[10] with increasing helium partial pressure when the helium was added through the CO_2 port and CO_2-He mixed with the vibrationally excited N_2 in the interaction region. An even greater increase in power output, by roughly a factor of two, was observed by Patel when the helium was added through the N_2 port and was thus in the parallel discharge with the N_2 prior to mixing with CO_2 in the interaction region. This same effect has been observed with a parallel-pumped tube built at IBM and shown in Figure 3.

The electron-energy distribution in the positive column of a normal glow discharge is controlled in part by the ionization potentials of the components of the gas mixture. The trend is to a higher electron temperature with higher ionization potentials[4, 11]. The ionization potentials of N_2, CO_2 and He are 15.5, 13.79 and 24.58ev, respectively. Typical mixtures used for laser emission vary, but a mixture of 2/1/10, N_2/CO_2/He is not uncommon.

Thus, there is some evidence that one of the significant roles played by helium in N_2-CO_2-He lasers is to shift the electron temperature to a slightly higher value for a given total pressure and thus increase the pumping of the upper laser level by direct electron impact and by vibrational eneryg transfer from $N_2^*(v=1)$.

The pulsed plasma tube used for most of the H_2-C_2H_2-He work is straightforward and is shown in Figure 1. The mirrors are exposed to the vacuum, and are mounted by means of stainless steel bellows. Operation of the tube with N_2-CO_2-He is indicated in Figure 2. The ratio of N_2:CO_2:He used for the data shown was 2:1:4 at 8 Torr and produced pulse energies of 15 to 20 millijoules.

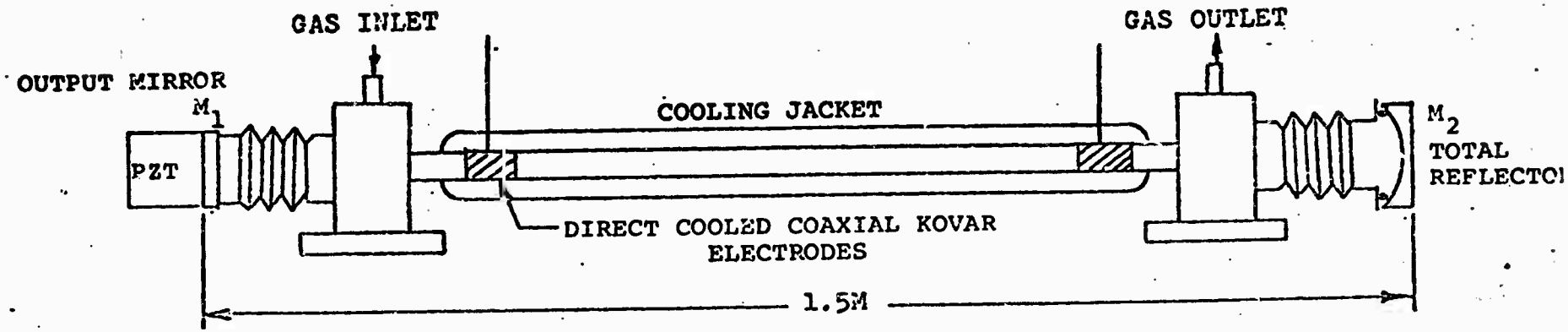
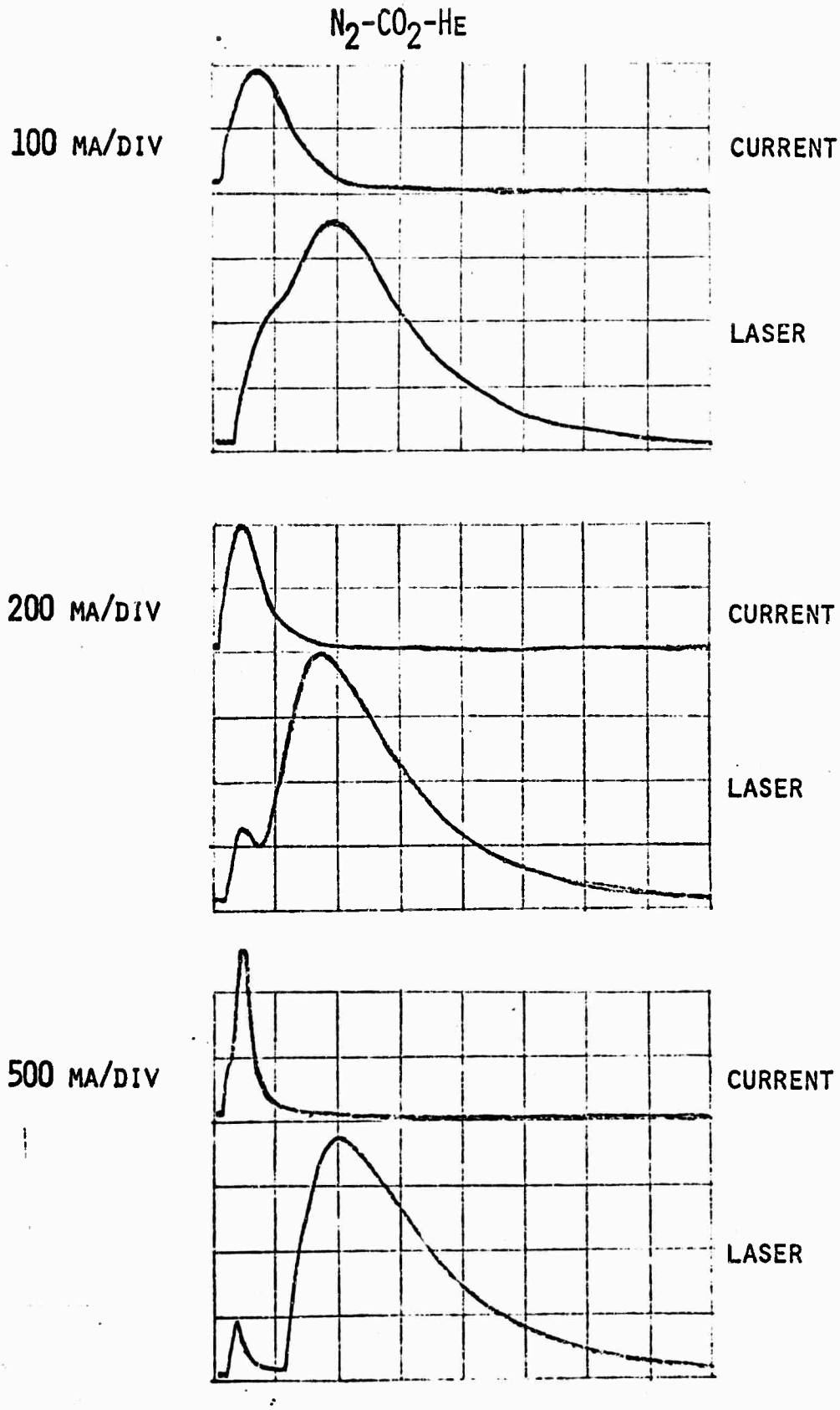


Figure 1. Pulsed Excitation Plasma Tube with Coaxial Kovar Electrodes.



57.1% He
28.6% N_2
14.3% CO_2
P = 8 Torr

Figure 2

The parallel tube shown in Figure 3 produced CW power outputs in excess of 600mW from CO_2 which had been pumped only from a resonant energy transfer from N_2 which was excited in the parallel discharge. For these experiments a 95% reflectivity dielectric mirror was used for output coupling, and an input power of 220 watts was used. This compares to an output power of the order of 2mW with an input power of 100 watts obtained by Patel[5].

This parallel tube was also operated in the pulsed mode by the addition of two electrodes near the center of the discharge region to prevent the plasma from traveling down the interaction region. Figure 4 shows a diagram of the modified tube and the pulse circuitry.

A 0-16Kv, 0-12ma dc power supply was used to charge a 0.02uf capacitor. This charge was then applied to the plasma tube when the electrode shown on the right in Figure 4 was switched to ground by the thyratron tube. The pulse repetition rate could be controlled by a pulse generator which triggered the thyratron. Two Welch Model 1397, 15cfm each, mechanical pumps were operated in parallel in order to continuously flow gas mixtures through the plasma tube. Partial pressures and flow rates of up to three component gases could be controlled through the use of needle valves, flow meters and a manifold arrangement. Partial pressures were measured upstream of the plasma tube using a capacitor manometer.

Figure 5 shows the performance of this tube with CO_2 under pulsed conditions. Output pulse energies were typically 3 millijoules, and the peak power was approximately 7 watts. The nitrogen flow rate was approximately 60 liters/min at the tube which produced a flow velocity of 400cm/sec in each half of the interaction region. Morgan and Shiff have measured the lifetime of the metastable $N_2^*(v = 1)$ level at a few Torr to be 114msec, which means that if wall collisions are unimportant, the excited molecules travel 45cm in one decay time. Considering both halves of the tube, this represents almost the entire 1 meter length of the interaction region.

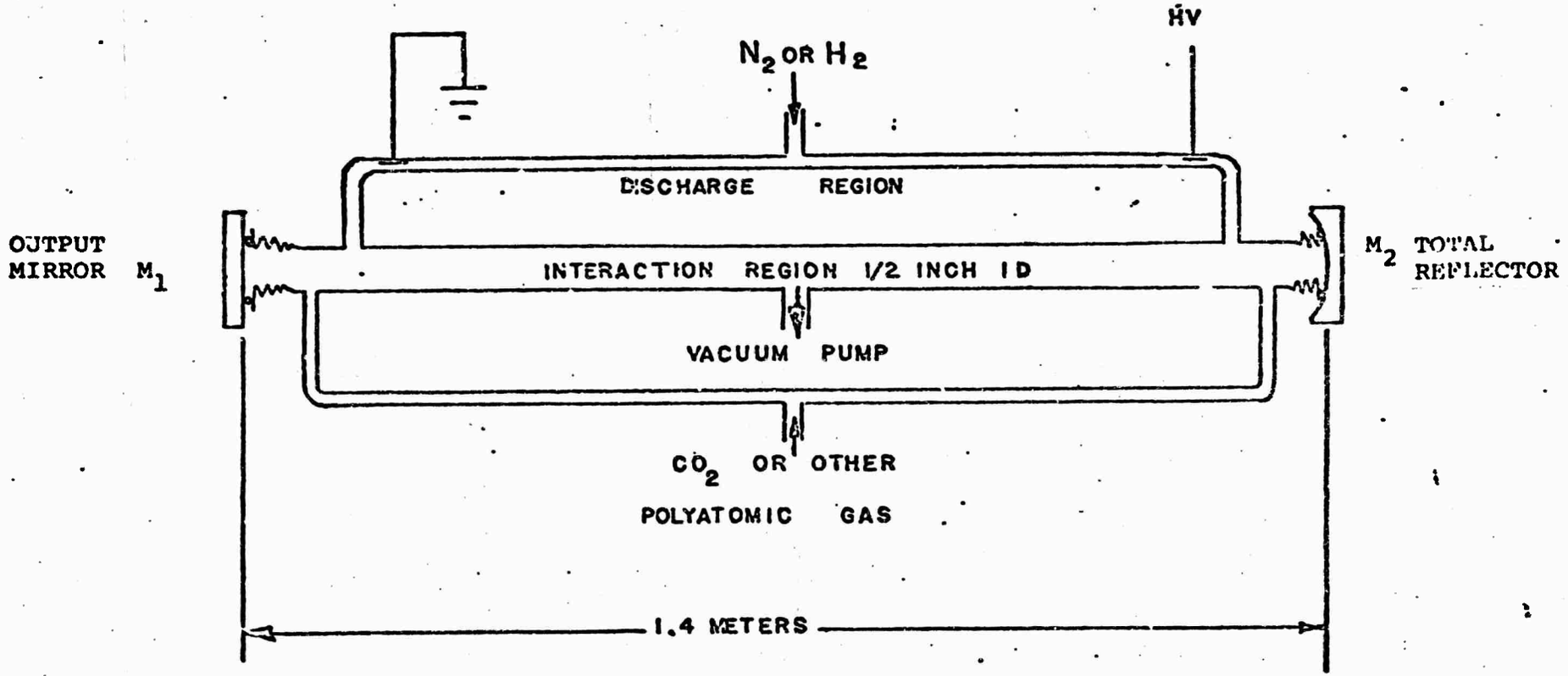


Figure 3. Parallel Tube

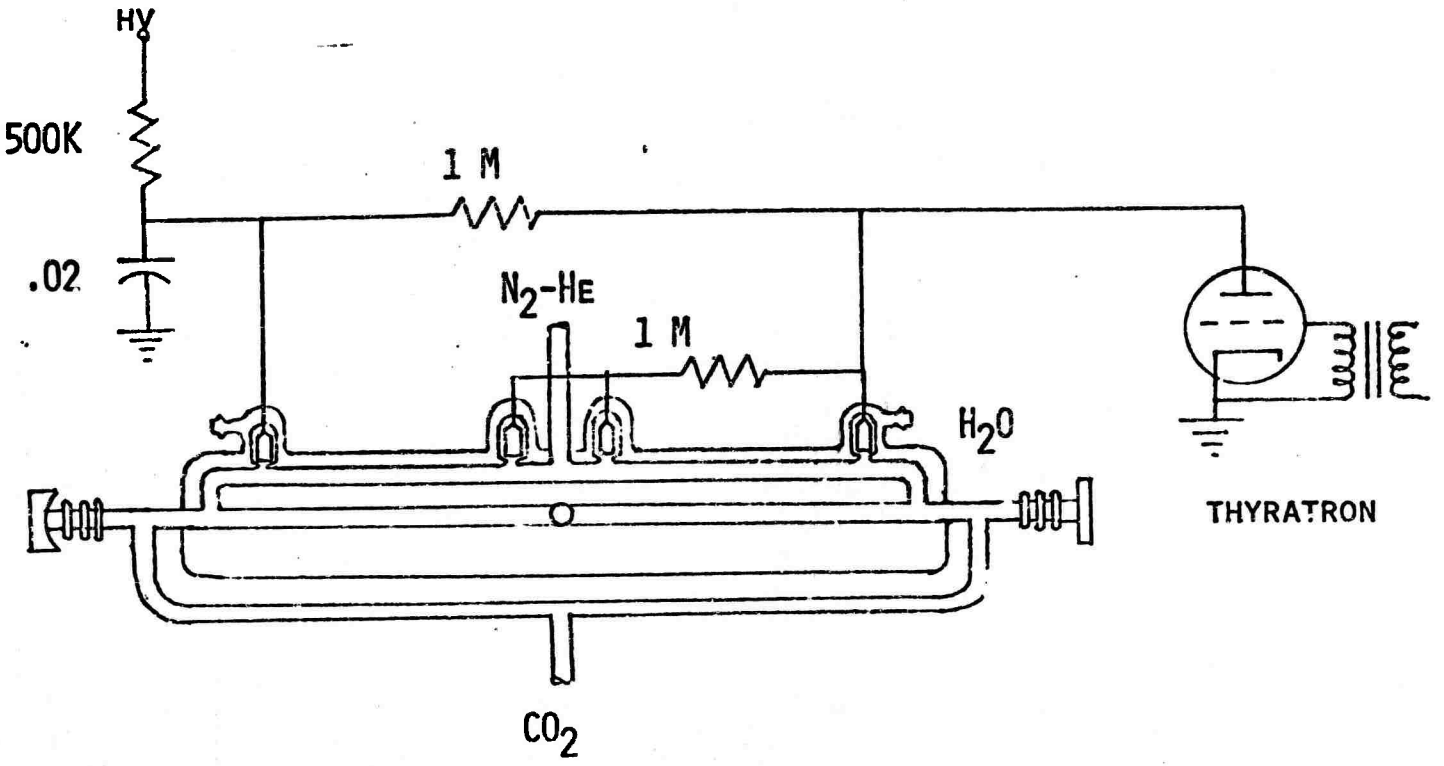


DIAGRAM OF PULSED PARALLEL TUBE

Figure 4

PULSED N_2-CO_2-HE IN PARALLEL TUBE

Figure 5

3.0 STIMULATED EMISSION FROM H_2-C_2H_2-He

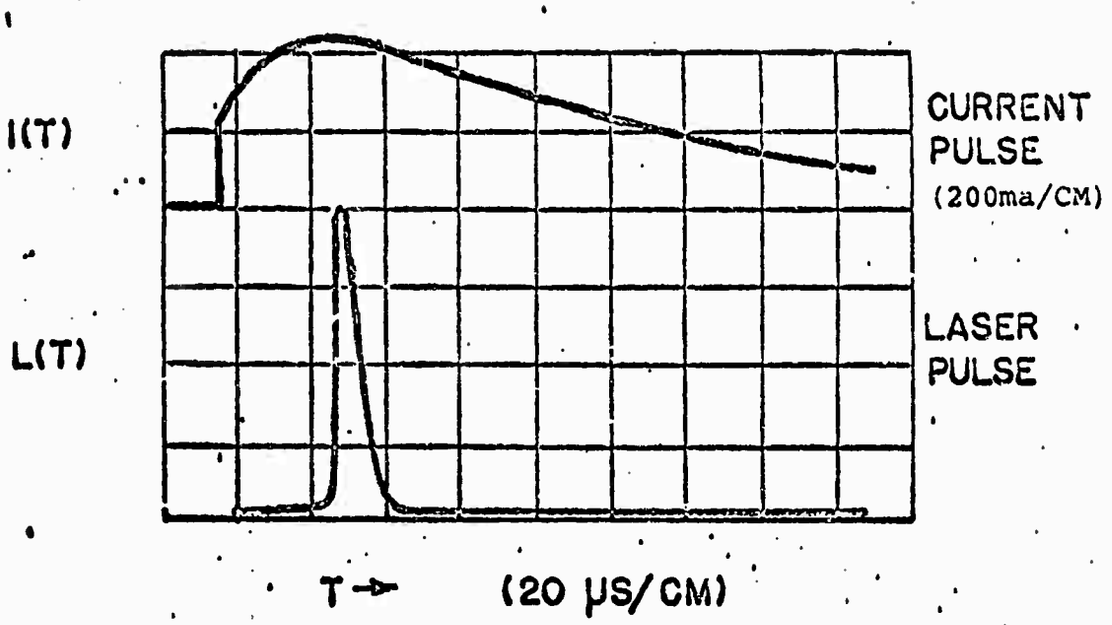
Laser emission near 8 microns has been obtained from a flowing mixture of H_2-C_2H_2-He under pulsed excitation conditions in a gas discharge[12.13]. This lasing action may result from vibrational excitation of C_2H_2 via a near resonant energy transfer from the metastable v = 1 vibrational level of H_2 which has been excited in the helium rich discharge as well as possible direct electron impact excitation of C_2H_2. Laser action has been observed, however, without the presence of hydrogen in the discharge.

A Jarrell-Ash one-meter Czerny-Turner spectrometer with a 98 groove/mm IR grating, blazed at 7μ was used to measure the wavelength of the laser emission. This grating gave a linear dispersion of approximately 102Å/mm and a theoretical resolving power of 8A in the first order. The precision of the wavelength counter is ±1A with a 1180 groove/mm grating. This corresponds to a precision of ±12A with the 98 groove/mm grating. A liquid nitrogen cooled Ge:Au detector was used in these experiments. This detector has a specified time response of 20ns

3.1 H_2-C_2H_2-He LASER CHARACTERISTICS

Representative results with the pulsed tube in Figure 1 showing the current pulse through the H_2-C_2H_2-He plasma and the laser pulse obtained are shown in Figures 6 and 7. A flowing mixture of approximately 1 torr C_2H_2, 2 torr H_2 and 20 torr He was used in each case. The values of peak powers given were measured with no attempt being made to optimize the output coupling.

The first results were obtained using an aperture for broadband output coupling and is shown in Figure 6. The laser emission was usually found to be on a single line at 8.040μ and no rotational structure was observed. However,



M_1: Flat Au-coated BaF_2 with 1/2 mm dia. hole

M_2: R=5 meters Au-coated

$H_2/C_2H_2/He$=2/1/20 torr

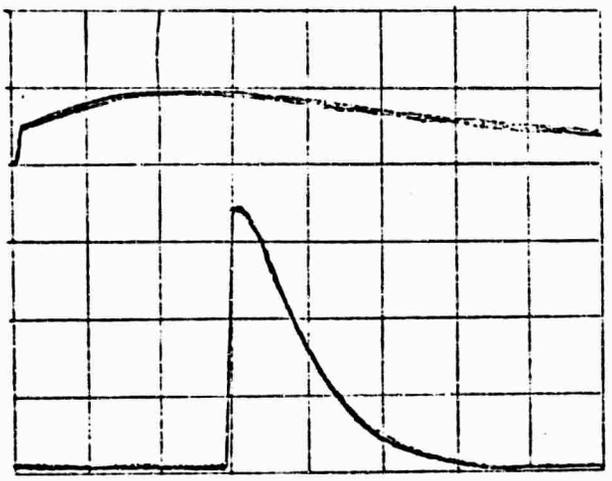
T_{jacket}=22°C

1/2 inch ID Plasma tube with neon sign electrodes
Peak power ≈ 5.7 watts
PRF = 32 pulses/sec

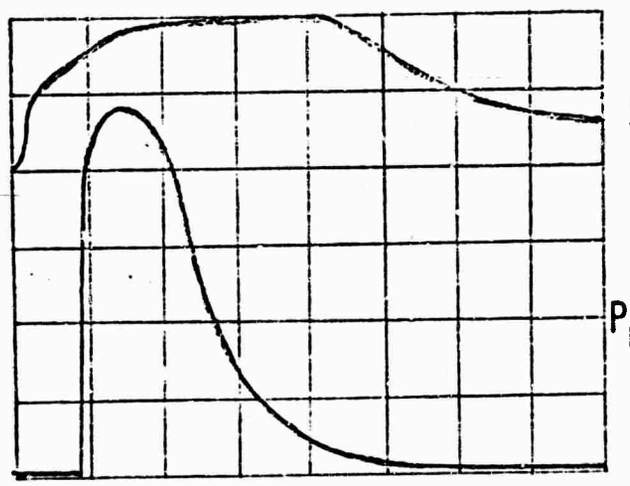
Figure 6. First Laser Output Obtained from H_2-C_2H_2-He Mixture.

H_2-C_2H_2-HE

2:1 20 Torr



25° C



-80° C

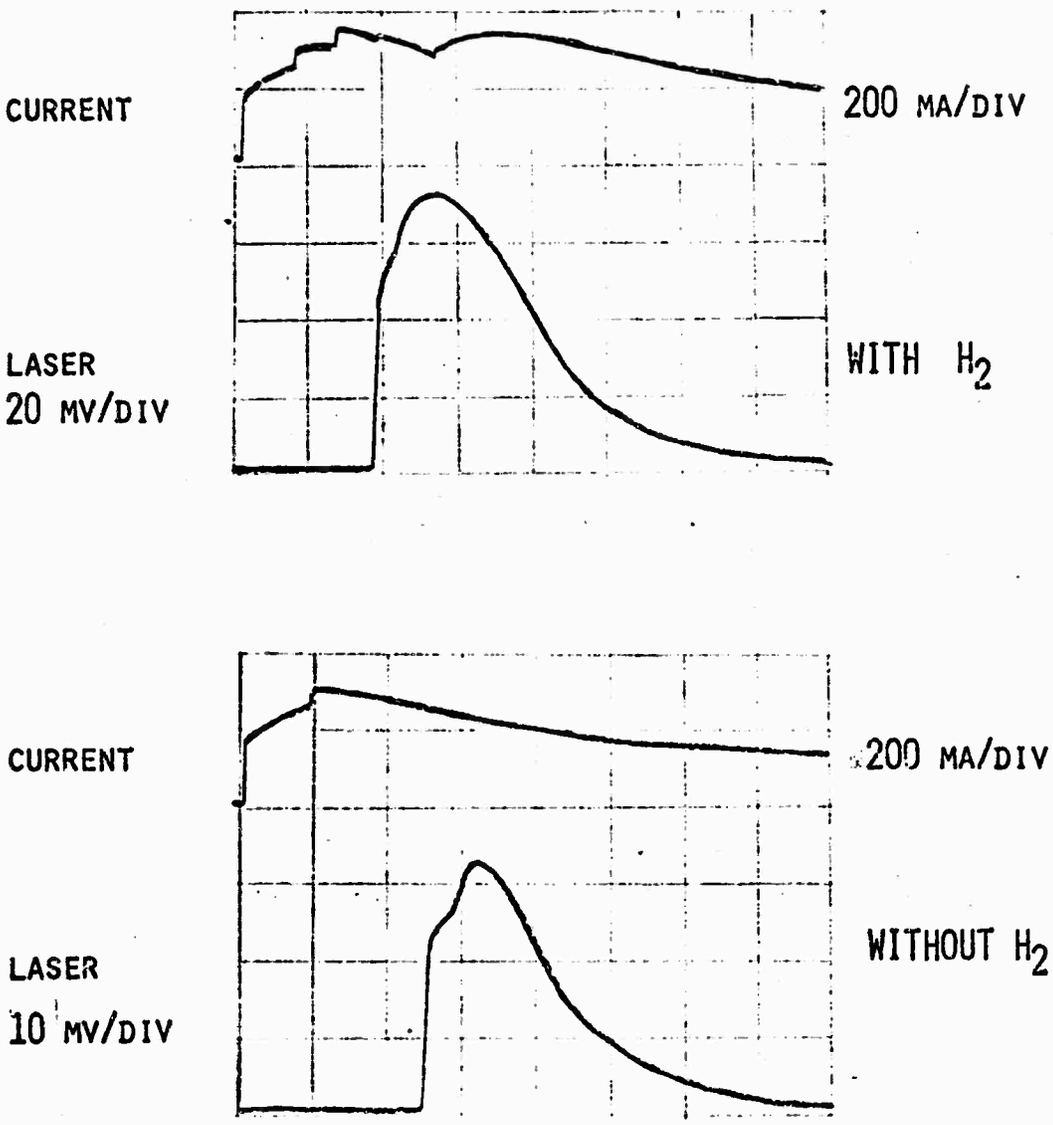
2:1:23 Torr

Figure 7

on one occasion, two lines were observed at 8.034 μ and at 8.040 μ.

The results obtained using narrow band dielectric mirrors are shown in Figure 7. The laser pulses shown are for two cooling jacket temperatures. Figure 7b shows the result obtained when the gas mixture was pre-cooled and the cooling jacket temperature was -80°C. Five emission lines were observed at different times under these conditions. The wavelengths in air of these five lines were measured as 8.0313, 8.0329, 8.0352, 8.0383 and 8.0416 μ using the 12th order of the 6678Å and 6717Å neon lines as a calibration reference. The accuracy of these wavelength measurements was probably better than ±2A. A 40:1 increase in peak power was observed in cooling the discharge from 22°C to -80°C as noted in Figure 7.

Laser action in H_2-C_2H_2-He in the 1/2 inch I.D. plasma tube could only be obtained over a very narrow range of discharge conditions, i.e., discharge current, and over a very narrow range of gas mixtures. Laser emission was not observed from a pure C_2H_2 plasma or a C_2H_2-H_2 plasma, but it was observed with a C_2H_2-He plasma. Figure 8 shows the results when the hydrogen was removed from the discharge. In the upper curve, the pressures of H_2-C_2H_2-He were 2, 1, and 32 torr, respectively, and in the lower curve the pressures were 0, 1, 32 torr. In both cases, both the gas and cooling jacket were at -60°C. Without hydrogen, the peak power was reduced by a factor of 2, and the pulse energy was reduced even more because of the slight decrease in pulse width. The same spectral lines were observed in the laser output both with and without hydrogen.



OPERATION WITHOUT HYDROGEN

Figure 8

Lasing again with reduced output occurred when N_2 was substituted for H_2. In this case an increasing delay time ranging from 20 to 65 usec accompanied the decreasing output.

These last results indicate that either some species other than C_2H_2 may be producing the laser emission or that direct electron impact excitation of C_2H_2 can alone lead to laser emission. They do not, however, completely rule out resonant energy transfer from $H_2^*(v = 1)$ as a possible pumping mechanism.

These results further support the fact that much work is needed in understanding the complex discharge chemistry and various pumping mechanisms involved as discussed in Section 3.3.

Laser emission was obtained at pulse repetition rates from 1pps up to 250 pps. Power supply limitations, i.e., current capability, prevented operation at higher repetition rates. The discharge ran quite clean, although light carbon deposits were formed near the electrodes after many hours of operation at pulse repetition rates of the order of 26pps. The side light emission from the plasma was very weak during laser action and, in fact, could not be observed with the eye when the laboratory lights were turned on.

3.2 POSSIBLE IDENTIFICATION OF LASER TRANSITIONS

Acetylene, C_2H_2, is a linear symmetric molecule with five normal modes of vibration[14]. These normal modes are shown in Figure 9. Some of the vibrational energy levels of the ground electronic state of C_2H_2 are shown in Figure 10. In contrast to the $\nu_3 - \nu_1$ band of CO_2, the $\nu_2 - \nu_5^1$ ($01^10 - 00^01^0$) band of C_2H_2 has an allowed Q-branch[14]. This $\nu_2 - \nu_5^1$ band Q-branch has been observed in absorption by Bell and Nielsen[24,25] with

C_2H_2 - ACETYLENE

Linear Symmetric Molecule

H-C≡C-H

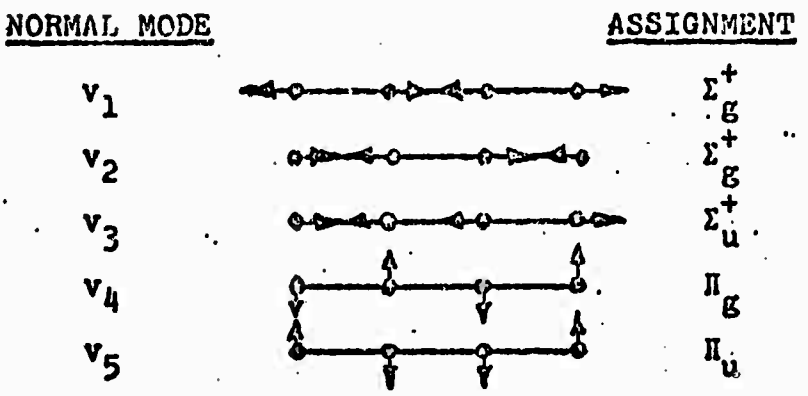


Figure 9. Normal Vibrational Modes of Acetylene

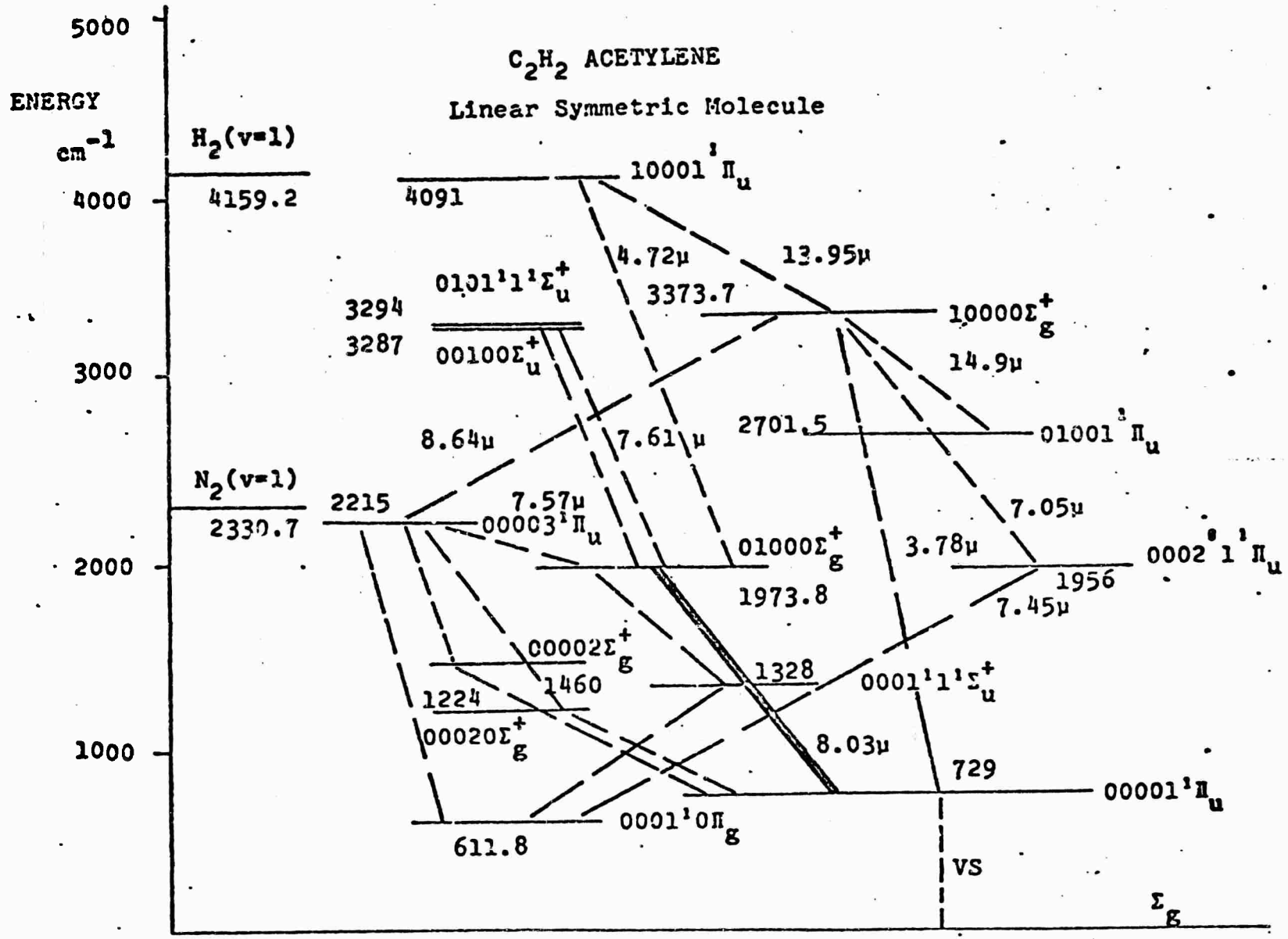


Figure 10. Vibrational Energy Level Diagram of Acetylene

Thus, equation (3-3) can be written as

$$Q(J) = \nu_o' + (B_U - B_L)J(J+1) + \ldots \qquad (3\text{-}4a)$$

This equation can be used to calculate the energies of the Q-branch lines of the $\nu_2 - \nu_5^1$ band of C_2H_2. The results obtained using Herzberg's[14] values of B_e, and the α_i's,

$B_e = 1.1838\text{cm}^{-1}$

$\nu_o' = 1245.28\text{cm}^{-1}$ *

$\alpha_1 = 0.008\text{cm}^{-1}$

$\alpha_2 = 0.0063\text{cm}^{-1}$

$\alpha_3 = 0.0056\text{cm}^{-1}$

$\alpha_4 = -0.0013\text{cm}^{-1}$

$\alpha_5 = -0.0022\text{cm}^{-1}$

giving,

$B_U = B_{\nu_2} = 1.17105\text{cm}^{-1}$

$B_L = B_{\nu_5^1} = 1.17955\text{cm}^{-1}$

or, $\Delta B = B_U - B_L = -0.00850\text{cm}^{-1}$

are shown in Table 1 for the first twenty Q-branch transitions of the $\nu_2 - \nu_5^1$ band of C_2H_2. Normally, for the C_2H_2 the lines with odd J should be more intense than the even J due to the 3:1 ratio of the statistical weights[14].

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* Herzberg gives a value for $\nu_o' = 1244.7\text{cm}^{-1}$. We have adjusted this value by 0.58cm^{-1} to give a better fit to the experimentally determined wavelengths.

TABLE I

Q-Branch Lines of the $\nu_2 - \nu_5'$ Band of C_2H_2 Calculated from Herzberg's Data

J	ΔE	$\lambda_{calc.}$	$\lambda_{obs.}$
1	1245.26	8.0282	
2	1245.23	8.0285	
3	1245.18	8.0288	
4	1245.11	8.0292	
5	1245.03	8.0298	
6	1244.92	8.0304	
7	1244.80	8.0312	8.0313*
8	1244.67	8.0321	
9	1244.52	8.0331	8.0329
10	1244.35	8.0342	
11	1244.16	8.0354	8.0352
12	1243.95	8.0367	
13	1243.73	8.0381	8.0383
14	1243.50	8.0397	
15	1243.24	8.0413	8.0416
16	1242.97	8.0431	
17	1242.68	8.0449	
18	1242.37	8.0469	
19	1242.05	8.0490	
20	1241.71	8.0512	

*This line observed only once.

The results obtained Keller's[15] values of B_e, and the α_i's,

$B_e = 1.1845 cm^{-1}$

$\nu_o' = 1245.20 cm^{-1}$ **

$\alpha_1 = 0.0063 cm^{-1}$

$\alpha_2 = 0.0092 cm^{-1}$

$\alpha_3 = 0.0053 cm^{-1}$

$\alpha_4 = 0.00065 cm^{-1}$

$\alpha_5 = 0.0021 cm^{-1}$

giving,

$B_U = 1.1673 cm^{-1}$

$B_L = 1.1786 cm^{-1}$

or,

$$B = B_U - B_L = -0.0113 cm^{-1}$$

are shown in Table 2 for the first twenty Q-branch transitions of the $\nu_2 - \nu_5^1$ band of C_2H_2. All values of ΔE are given for vacuum and wavelengths are given in air.

** Keller's value for $\nu_o' = 1244.79 cm^{-1}$ which is an adjustment of $0.41 cm^{-1}$.

TABLE 2

Q-Branch Lines of the $\nu_2 - \nu_5'$ Band of C_2H_2 Calculated from Keller's Data

J	ΔE	$\lambda_{calc.}$	$\lambda_{obs.}$
1	1245.18	8.0288	
2	1245.13	8.0291	
3	1245.06	8.0295	
4	1244.97	8.0301	
5	1244.86	8.0308	8.0313*
6	1244.73	8.0318	
7	1244.57	8.0327	8.0329
8	1244.39	8.0339	
9	1244.18	8.0352	8.0352
10	1243.96	8.0367	
11	1243.71	8.038[illegible]	8.0383
12	1243.44	8.0400	
13	1243.14	8.0419	8.0416
14	1243.82	8.0440	
15	1242.49	8.0462	
16	1242.13	8.0485	
17	1241.74	8.0510	
18	1241.34	8.0536	
19	1240.91	8.0564	
20	1240.45	8.0594	

*This line only observed once.

A piezoelectric drive was used to scan one laser mirror in order to average de pulling effects and assure mode coincidence with any rotational line having sufficient gain to lase. An indication of these effects are that while operating the drive we have seldom found that a rotational line is missing. This is commonly the case with a fixed cavity length. Thus, scanning the cavity provides the best (unshifted) lasing spectrum available.

Phase sensitive synchronous amplification of a Ge:Au detector output was used; the amplifier gives a dc output proportional to the laser emission which is used as input to the Y-channel of an X-Y recorder. A potentiometer mounted to the wavelength drive of the spectrometer gave a dc voltage proportional to the wavelength which then drives the X-channel of the recorder. An additional moveable mirror and an S-1 PMT is used to enable the 12th order of the Ne calibration lines to be recorded on the X-Y recorder along with the C_2H_2 laser lines. In all cases, dielectric mirrors were used. The total reflector has a 5 meter radius and a reflectivity of 99.5% $\pm$0.5% at 8u. The output mirror is flat with a reflectivity of 98% $\pm$5% at 8u.

Figure 11 shows a typical spectrum, and, in addition, shows the calculated wavelength of the rotational lines in the Q-branch for the rotational constants of Herzberg[14] and Keller[15]. The best fit is seen using the rotational constants of Keller. Although this fit is quite reasonable, all doubts about the identification of the transition should be removed by additional experiments, on absorption in C_2H_2 planned for the third quarter.

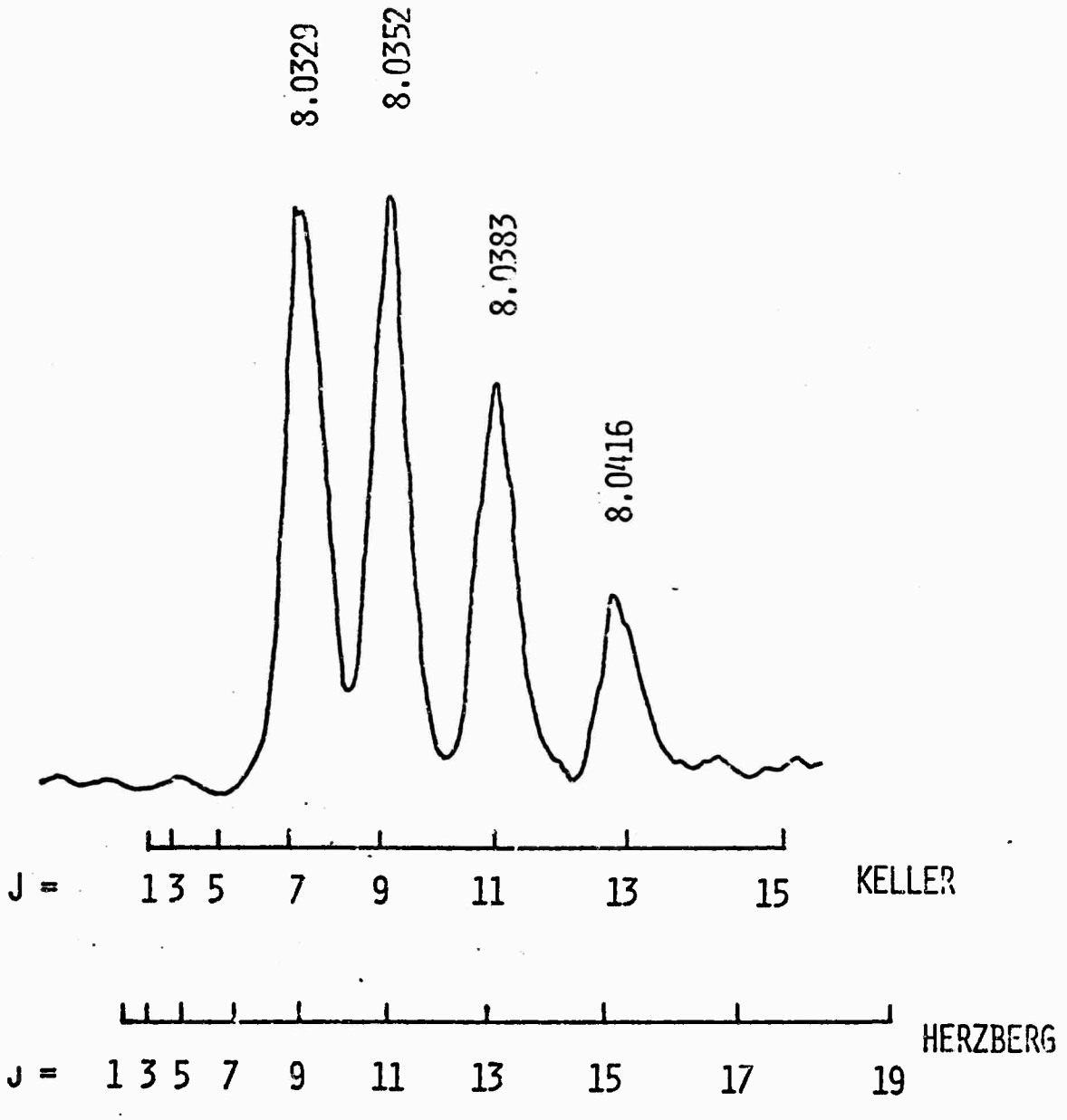
8.0329
8.0352
8.0383
8.0416
J = 1 3 5 7 9 11 13 15
KELLER
J = 1 3 5 7 9 11 13 15 17 19
HERZBERG

Figure 11

It is also possible that the laser emission is originating from transitions in some other species which is a dissociative product of the discharge. The carbon formation around the electrodes indicates that some dissociation of the C_2H_2 is occurring to form free carbon. The dissociation energies of C_2H_2 are given by Lathan, et al[23]. The pertinent reactions are

$$C_2H_2 \rightarrow C_2H + H \qquad (4.97ev)$$

$$C_2H \rightarrow C_2 + H \qquad (5.76ev)$$

$$C_2 \rightarrow 2C \qquad (6.25ev)$$

$$C_2H_2 \rightarrow 2CH \qquad (9.95ev)$$

These values are the same order of magnitude as the $CO_2 \rightarrow CO + O$ band strength at 5.6ev. Thus, there are certainly many dissociative products of C_2H_2 in the discharge.

An observation of the visible spectrum of the discharge showed fairly weak CH lines, C_2 Swan bands, H_β and He emission lines. None of these observed bands or lines are very strong, including the C_2 bands, indicating that the dissociation was not appreciable.

Ethylene, C_2H_4, and ethane, C_2H_6, could also be formed in the discharge through hydrogenation of C_2H_2.

$$C_2H_2 + H_2 \rightarrow C_2H_4$$

$$C_2H_4 + H_2 \rightarrow C_2H_6$$

It is also possible that formation of diacetylene, C_4H_2, can occur.

Thus, possible constituents of the plasma include C_2H_2, C_2H_4, C_2H_6, C_4H_2, CH, CH_2, CH_3 and C_2H_5 as well as ions of these species and the laser emission may be originating from one of these.

The observation of only five emission lines from the laser, probably due to rapid rotational thermalization caused by the high helium content of the plasma, makes a positive identification of the laser transitions from rotational structure difficult.

3.3 DISCUSSION OF POSSIBLE PUMPING MECHANISMS

Assuming that the laser emission is occurring on Q-branch transitions of the C_2H_2 $\nu_2 - \nu_5^1$ band, the laser would then be functioning as a classical four level laser (refer again to Figure 10). Two possible pumping mechanisms for obtaining population inversion in C_2H_2 are direct electron impact excitation and a near resonant vibrational energy transfer from the $H_2^*(v = 1)$ which has been excited by electron impact in the helium rich plasma.

$$C_2H_2 + e \rightarrow C_2H_2^* + e \qquad (3\text{-}5)$$

$$H_2(v = o) + e \rightarrow H_2^*(v = 1) + e \qquad (3\text{-}6)$$

$$H_2^*(v = 1) + C_2H_2 \rightarrow H_2(v = o) + C_2H_2^*(\nu_1 + \nu_5^1) + \Delta E = 68.2\text{cm}^{-1} \qquad (3\text{-}7)$$

The excited state produced by the inelastic collision process shown in Equation (4-5) is not indicated since the details of this process are now known at this time, nor is the relative importance of this mechanism in pumping C_2H_2 known. The $C_2H_2^*(\nu_1 + \nu_5^1)$ level on the right-hand side of process (3-7) can cascade via collisions or radiative transitions to the ν_2 level, which is the upper laser level. The lower laser level, the ν_5^1 level, can be de-populated through the strongly allowed ν_5^1 to the ground vibration level transition.

Vibrationally excited, ground-electronic-state hydrogen molecules have been observed as a long-lived product of a microwave discharge in pure hydrogen gas by Heidner and Kasper[16]. These excited molecules were identified by their vacuum-ultraviolet absorption spectrum. They concluded that,

at a pressure of 3 torr, hydrogen passed through a microwave discharge contains approximately 1-4% of the molecules in the $v'' = 1$ state 25msec after leaving the discharge. This compares with 30% of the N_2 molecules in the $v'' = 1$ state in a low pressure microwave discharge in pure N_2 as determined by Kaufman and Kelso[17].

The relaxation time of the $H_2^*(v = 1)$ level should be about 250msec at $T = 300^oK$ and $p = 3$ torr[16, 18]. This compares to the relaxation time of 114msec of $N_2^*(v = 1)$ at $T = 300^oK$ and pressures of the order of a few torr as determined by Morgan and Schiff[24].

The cross section as a function of electron energy, $\sigma(E)$ for the process (3-6) has been measured by Schulz[19]. It has a peak value of $0.55 \times 10^{-16} cm^2$ at 2.2ev compared to a peak value of $1.5 \times 10^{-16} cm^2$ at 2.2ev for the vibrational excitation of N_2 by direct electron impact. From $\sigma(E)$, the rate coefficient for the excitation of H_2 by direct electron impact can be calculated[20].

$$X(t) = N_e(t) \langle \sigma v \rangle \tag{3-8}$$

where $N_e(t)$ is the electron density in the plasma and $\langle \sigma v \rangle$ is Schulz's cross section averaged over the electron velocity distribution in the plasma. Assuming a Maxwellian energy distribution for the electron in the plasma,

$$\langle \sigma v \rangle = K_o T_e^{-3/2} \int E \quad \sigma(E) e^{-E/T_e} dE \tag{3-9}$$

where $K_o = 6.6971 \times 10^7$ and T_e is the average electron temperature in ev. The term $\langle \sigma v \rangle$ given by Equation (3-9) is plotted versus electron temperature in Figure 12. It is clear from Equations (3-8) and (3-9) and from Figure 12

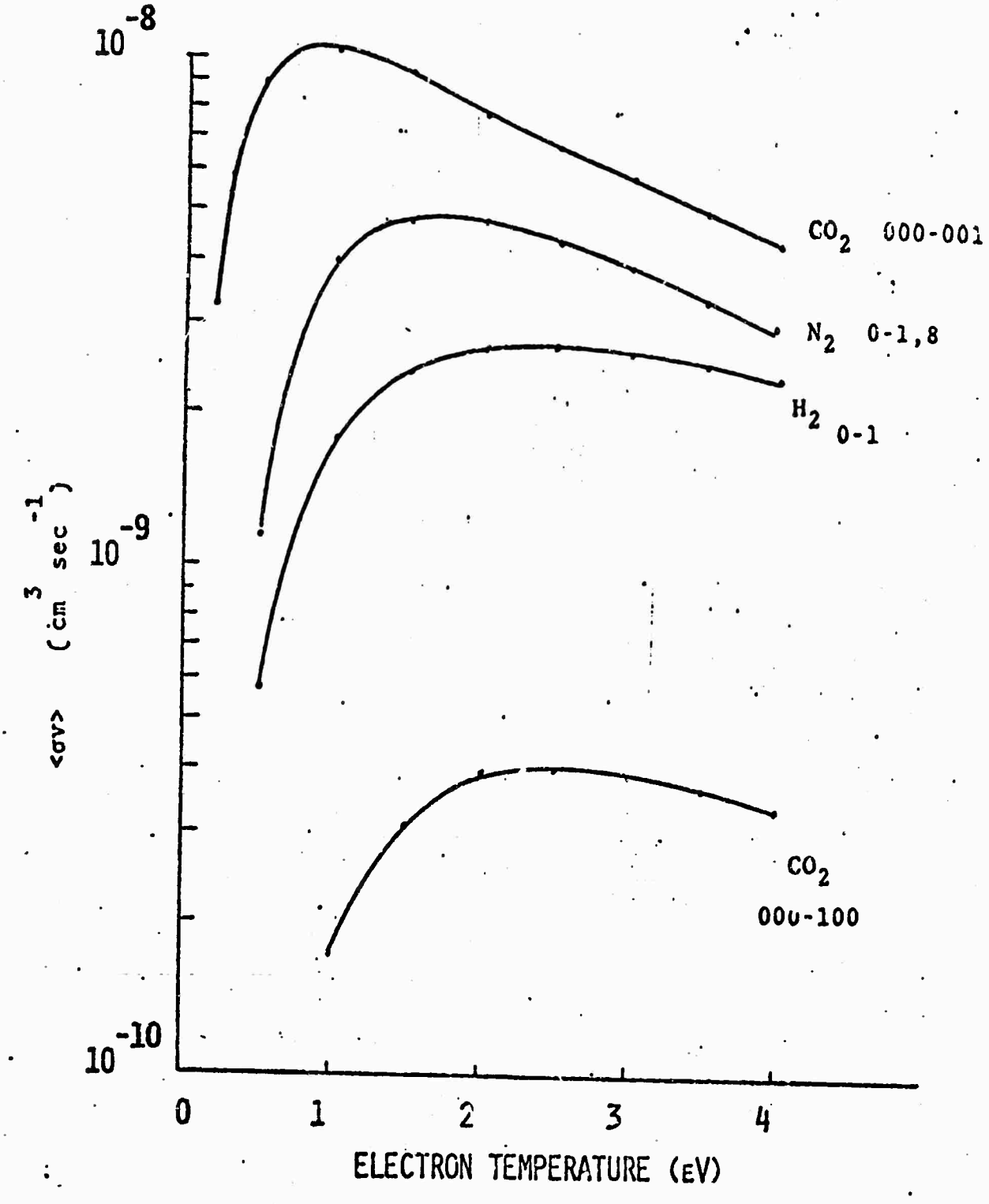

Figure 12. <σv> vs. Electron Temperature

that the rate coefficient for vibrational excitation of H_2 by direct electron impact depends both upon the electron temperature and the electron density in the plasma.

Data is available in the literature on the electron temperature in pure gases under normal glow discharge conditions, but very little data is available for gas mixtures. The electron temperature, T_e, as a function of ionization potential, u_i, pressure, p, and tube radius, R, and an empirical constant, c, which depends on the gas, is given by Brown[11] (see also von Engel[12], pages 63 and 242).

$$\left(\frac{U_i}{T_e}\right)^{-1/2} \exp\left\{\frac{U_i}{T_e}\right\} = 1.16 \times 10^7 c^2 p^2 R^2 \qquad (3\text{-}10)$$

The derivation of this equation is also presented in Reference 12. The electron temperature versus pressure in the positive column of a low pressure gas discharge in pure N_2, pure H_2 and pure He calculated from Equation (3-10) is shown in Figure 13. This equation only considers the ionization potential of the gas in determining the electron energy.

The source of electrons in a glow discharge is from ionization of the component gases and from secondary emission from the cathode. The energy of the electrons is controlled by elastic and inelastic collisions with the component gases. The inelastic processes are ionization and electronic excitations of all the components of the plasma, and vibrational and rotational excitations of the molecular components.

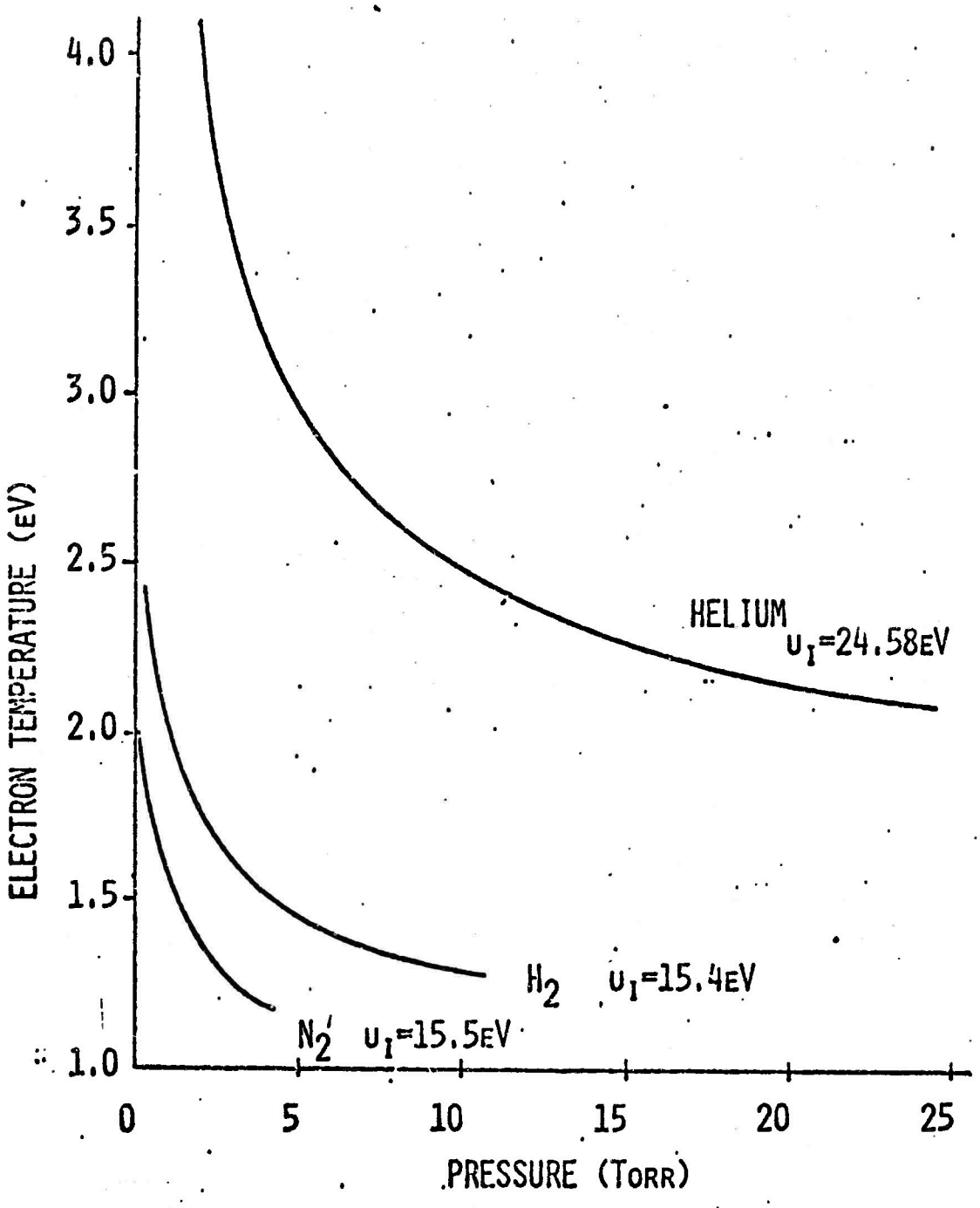

Figure 13. Electron Temperature vs. Pressure in Pure Gases

Again considering only the effect of the ionization potential, it can be shown that the electron temperature in a mixture of H_2-He can be somewhat higher than for H_2 alone at the same total pressure[12]. This same argument can be applied to H_2-C_2H_2-He and C_2H_2-he mixtures. This implies that high He partial pressures can be used to stabilize the discharge and still maintain high electron temperatures.

The use of pulsed excitation for H_2-C_2H_2-He also increases the excitation rate through the higher current density obtainable, by at least an order of magnitude, compared to dc operation with the gas mixture used.

3.4 PARALLEL PUMPING EXPERIMENTS

We conducted experiments with the parallel tube shown in Figure 4 to test the hypothesis of vibrational energy transfer from excited hydrogen to acetylene. A pulsed discharge in mixtures of hydrogen and helium occurred in the side-arm or discharge region of the tube. The gases then flowed into the interaction region of the tube where they were mixed with acetylene at room temperature. In each experiment, the mirrors were aligned by filling the tube with N_2-CO_2-He and adjusting for maximum average power.

A wide range of gas mixtures, flow rates, and total pressures were tried, and estimates indicate that hydrogen flow velocities up to 20 meters/sec were present. In all cases, the results were negative, and we have not observed laser emission from C_2H_2 in the parallel discharge tube.

Two problems with this experiment immediately come to mind: 1) the lower level population in acetylene, and 2) vibrational deactivation by collisions with the wall. The lower level of the lasing transition is only 729cm^{-1} above the ground state of acetylene. Thus, at room temperature, the population of the lower laser level is 3% to 4% of the ground state population. Larger inversions are required than for the case of CO_2, and the unpumped parts of the interaction region may be a sufficiently large fraction of the total length so that the gain cannot exceed the losses.

Diffusion coefficients have been used to estimate the lifetime of vibrationally excited H_2 if wall collisions are the source of deactivation. The mutual diffusion coefficient, D_{12}, for a binary mixture of hard, elastic spheres is[22]

$$D_{12} = \frac{3}{8} \left[\frac{\pi kT}{2M_r} \right]^{1/2} \frac{1}{\pi N d_{12}}$$

where M_r is the reduced mass $\frac{M_1 M_2}{M_1 + M_2}$,

$N = N_1 + N_2$ is the total number of molecules, and

$d_{12} = \frac{d_1 + d_2}{2}$ is the average molecular diameter.

If each such collision results in a deactivation, the average lifetime of an excited specie in an infinite cylinder of radius, r_0, is

$$\tau = \frac{1}{D_{12}} \left\{ \frac{r_o}{2.405} \right\}^2$$

Table 3 shows the results of lifetime calculations using these equations. The first entry is representative of the conditions in the parallel tube which results in lifetime of 0.732msec. Thus, even at the highest estimated flow velocity for hydrogen (2.0cm/msec), the excited molecules would only travel several centimeters before a very large fraction were deactivated. In our tube, the molecules have to travel approximately 10cm to reach the interaction region, so if wall collisions are effective it is unlikely that significant numbers of vibrationally excited hydrogen are available to pump the acetylene.

The second and third entries in the table represent the conditions in the simpler, pulsed tube, and the calculations show lifetimes 3 to 4 times greater than those expected in the parallel tube. The last entry is representative

TABLE 3

LIFETIME BY DIFFUSION TO WALLS

Pressures (Torr)			Temp (^{o}K)	D_{12} (cm^2/sec)	τ (msec.)
H_2	He	N_2			
5	5		300	95.2	0.732
2	28		300	31.7	2.196
2	28		193	25.5	2.738
	5	5	300	40.6	1.719

$d_{H_2} = 2.74$ Å

$d_{He} = 2.18$ Å

$d_{N_2} = 3.75$ Å

$M_{H_2} = 2.016$

$M_{He} = 4.002$

$M_{N_2} = 28.2$

$r_o = 0.635$ cm

of the situation of N_2-CO_2-He in the parallel tube. Based strictly on this lifetime of 2.2msec one would come to the same conclusions reached above and predict that CO_2 would not work in the parallel tube. This is obviously an erroneous conclusion as evidenced by the pulsed and CW operation reported in Section 2. Thus, some of the assumptions used in the calculation may be incorrect, at least for the N_2-He case. The Lewis-Rayleigh afterflow, which is a good indication of excited molecular nitrogen, can be seen throughout the interaction region and even some distance from the laser in the glass tubing leading to the vacuum pumps. In addition, the effectiveness of deactivating collisions with He has not been ascertained, and the small molecular weight of H_2 in contrast to N_2 may have important implications to the pumping process.

4.0 FUTURE PLANS

In an effort to further substantiate our identification of the lasing transition, we are planning an intracavity absorption experiment. We will test the ability of normal acetylene to quench the laser, thereby determining whether or not the lasing species is, in fact, acetylene or a discharge product.

We are considering the substitution of neon for helium to see if a heavier buffer gas affects the laser performance. In addition, we hope to further define the role of hydrogen by monitoring the visible emission from discharge products both with and without the presence of H_2.

Considering the calculated lifetimes for wall deactivation, estimated flow rates, and the presence of absorption from the lower level population, the parallel pumping experiments have been discontinued until after the absorption measurements are completed. They may be taken up again later with modified equipment.

A new plasma tube will be designed, built, and tested which will minimize the regions containing unexcited gases. Other gases will be investigated with this new tube during the latter portion of the contract period.

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