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Third Quarter, Technical Summary Report on
Molecular Laser Study

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INTRODUCTION

This third quarter Summary Technical Status Report is in partial fulfillment of the contract requirements as specified in the Contract Schedule, Section G, Paragraph I(D). The following sections present the Technical Report Summary as required in the Contract Schedule.

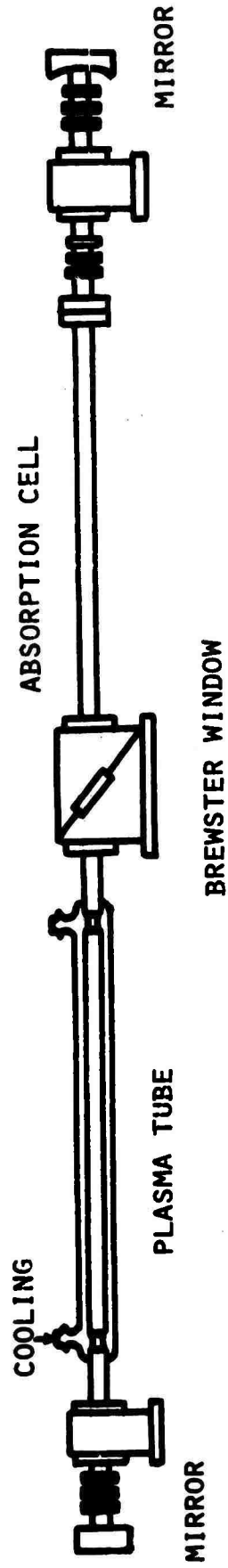
LASING TRANSITION

A good identification of the lasing transition was given in the last quarterly report based on the measured rotational spectra of the laser and the constants in Keller's thesis¹. In order to remove all doubts concerning the identification of the lasing species we performed an intra-cavity absorption experiment. The apparatus is indicated in Figure 1 and includes an additional gas cell inside the laser resonator. The plasma tube and the absorption cell are separated by a Brewster angle NaCl window. Only two rotational lines (Q(7) and Q(9)) were observed with this configuration even when a piezoelectric drive was used to scan one of the laser mirrors.

The laser spectra for three different C₂H₂ pressures in the absorption cell are shown in Figure 2. A pressure of 1.5 Torr was sufficient to prevent laser emission. However, laser emission was unaltered when the cell was opened to air at one atmosphere. Therefore, we conclude that molecular acetylene is definitely the lasing specie.

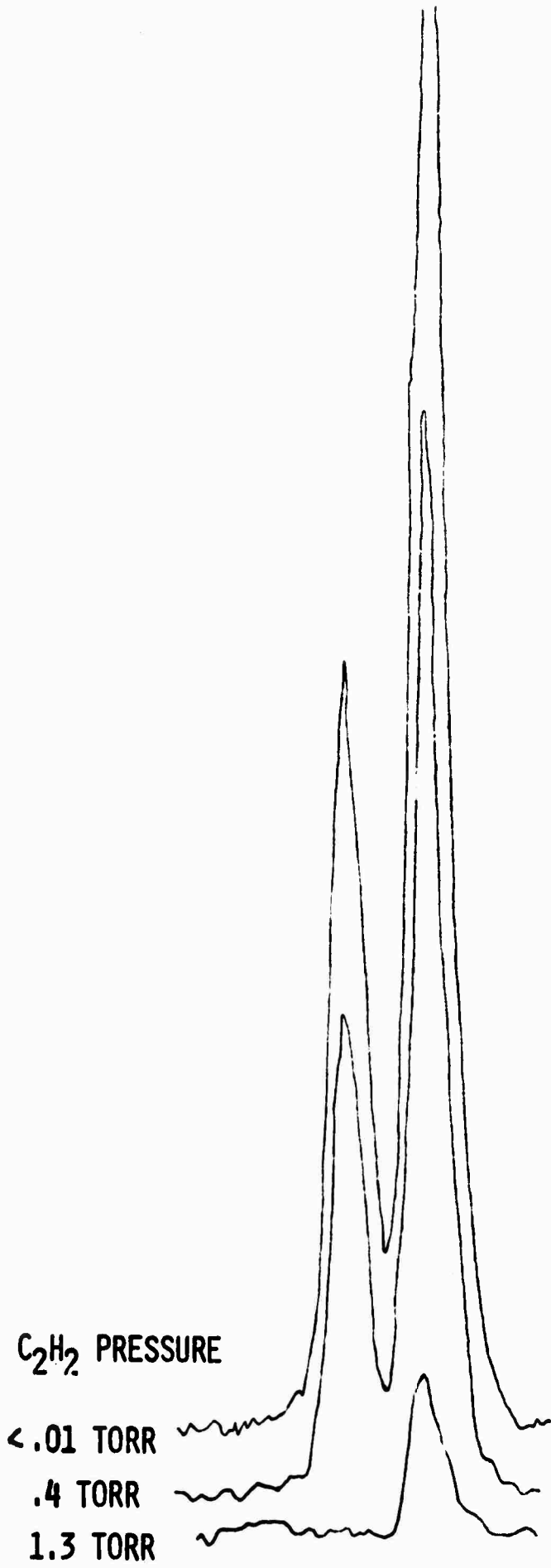
In an additional experiment, the absorption cell was placed outside the laser resonator, and the absorption was measured as a function of pressure and temperature. This data is shown in Figure 3. The measurements at the lower temperature were made with dry ice packed around the stainless steel absorption cell. A theoretical fit was made with the following expressions and is indicated by the solid curves:

$$D_J = \frac{C_1 P B_L e^{-729/kT} (2J+1) e^{-B_L J(J+1)/kT}}{T (T^{1/2} + C_2 P) (1 + e^{-619/kT} + e^{-729/kT})}$$



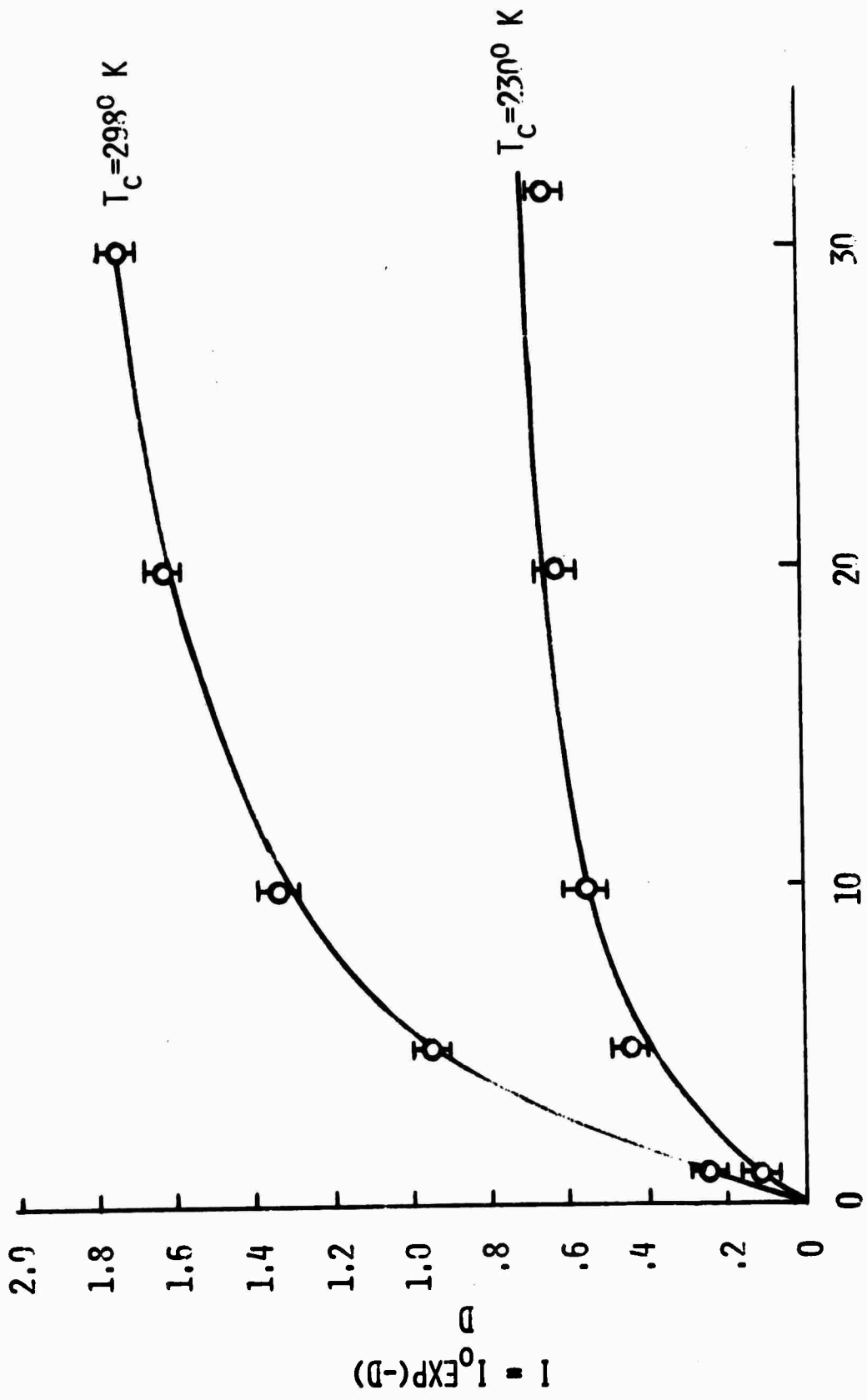
INTRACAVITY ABSORPTION APPARATUS

Figure 1



INTRACAVITY ABSORPTION EXPERIMENT

Figure 2



ACETYLENE PRESSURE (TORR)

ACETYLENE ABSORPTION IN EXTRACAVITY CELL
Figure 3

and

$$D = -\ln \frac{\sum W_J D_J}{\sum W_J}$$

where $J = 7, 9, 11, \text{ and } 13$; and W_J is a weighting factor to take the different intensities of the rotational lines into account. T is the temperature in K° , P is the pressure in Torr, k is Boltzmann's constant $= 0.6951 \text{ cm}^{-1}$, B_L is the rotational constant $= 1.1786 \text{ cm}^{-1}$, and $C_1 = 5300$ and $C_2 = 3$ are appropriate combinations of physical constants. The vibrational partition function includes the lowest two vibrational levels, and both Doppler and pressure broadening are taken into account. At room temperature pressure broadening dominates at pressures above 6 Torr. The constants, C_1 and C_2 were established by fitting the data at room temperature. In order to fit the low temperature results, a temperature $= 230^\circ K$ was required. This is 35° higher than the temperature of dry ice, but no measurement of temperature was made inside the cell, and small portions were not cooled at both ends. Thus, the accuracy of this temperature is not known.

The absorption measurements also provide an estimate of the laser gain. Based on an active length of 85cm and a total inactive length of 60cm in the plasma tube, and an acetylene pressure of 1.5 Torr in the intra-cavity absorption cell, one calculates a peak gain of 2.9db/meter.

EXCITATION MECHANISM

Further experiments were carried out ~~in the third quarter~~ to characterize the behavior of the laser in the absence and presence of H_2 . Data was taken in a new 1 inch I. D. plasma tube with coaxial electrodes, and the results are given in Table I. The flow rates for the C_2H_2 , H_2 , N_2 and He are given in terms of the centimeters of deflection on the flow meters, and the average power at 26Hz was measured with an Eppley thermopile. In all cases, the laser operated better with H_2 present in the discharge tube although the degree

Table I

EFFECTS OF H₂ AND N₂ ON THE C₂H₂ LASER

	<u>C₂H₂</u>	<u>H₂</u>	<u>N₂</u>	<u>He</u>	<u>T</u>	<u>Ave. Pwr.</u>
1.	.35	2.1	0	25+	-17°C	14 mW
2.	.35	0	0	25+	-18	10.5
3.	.35	0	2.1	25+	-17	2.1

4.	.9	2.1	0	25+	-16	19.6
5.	.9	0	0	25+	-17	11.2
6.	.9	0	2.1	25+	-17	-

7.	1.5	2.1	0	25+	-16	14
8.	1.5	0	0	25+	-16	0.28
9.	1.5	0	2.1	25+	-17	-

Total pressure in all cases was 32 Torr
Peak current was 1.75 amp
26 Hz rep.-rate

C₂H₂ - 0.35 cm 0.4 Torr
H₂ - 2.1 1.3 Torr
He - 25+ 30.3 Torr

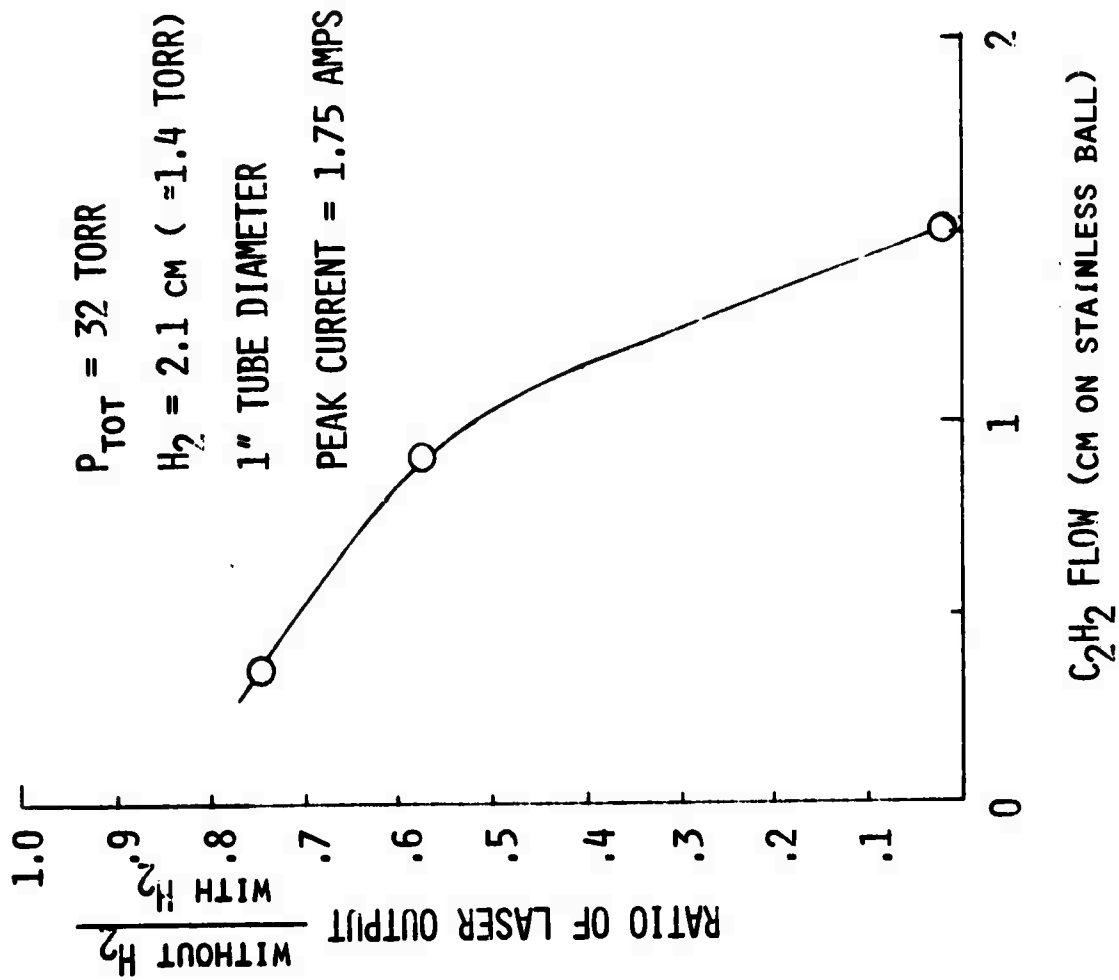
of improvement depends on the acetylene flow. This is shown in Figure 4. Thus, at higher acetylene flow rates the hydrogen is much more critical.

The substitution of N_2 for H_2 proves to be quite detrimental to the operation of the laser. With acetylene flow rates corresponding to .9 and 1.5cm the addition of N_2 caused the laser to cease operation.

The visible emission from the plasma tube was examined for identical discharge conditions to those indicated in Table I. Light was collected through a NaCl window on the cathode end of the plasma tube and monitored with a 1 meter spectrometer and a photomultiplier tube. Some of the results are shown below. Figure 5 is the spectra of the CH or methyne radical at 4314\AA with the flow rates corresponding to cases 1 and 2 in Table I. The intensity of the CH band is reduced by the addition of H_2 . Similar results were obtained for the C_2 Swan band at 5165\AA shown in Figure 6. Thus, the addition of H_2 to the C_2H_2/He discharge decreases the number of dissociative products from the acetylene. This also corresponds to the case of maximum laser output, and is consistent with the conclusion that acetylene and not a discharge product is the lasing species.

Figure 7 gives the spectra of the CH band for the substitution of N_2 for H_2 and corresponds to cases 2 and 3 in Table I. The intensity of the band is once again reduced. Note the appearance of the N_2^+ band at 4278\AA when N_2 is added.

There are several possible explanations for the observed behavior. First, since the laser operates without H_2 , direct impact excitation is obviously effective in populating the upper laser state. However, it is conceivable that this pumping mechanism is saturable, and resonant energy transfer from vibrationally excited hydrogen becomes more important at the higher acetylene pressures. This does not explain, however, the measurements of the intensities of the CH and C_2 bands.



EFFECT OF HYDROGEN ON C_2H_2 LASER OUTPUT

Figure 4

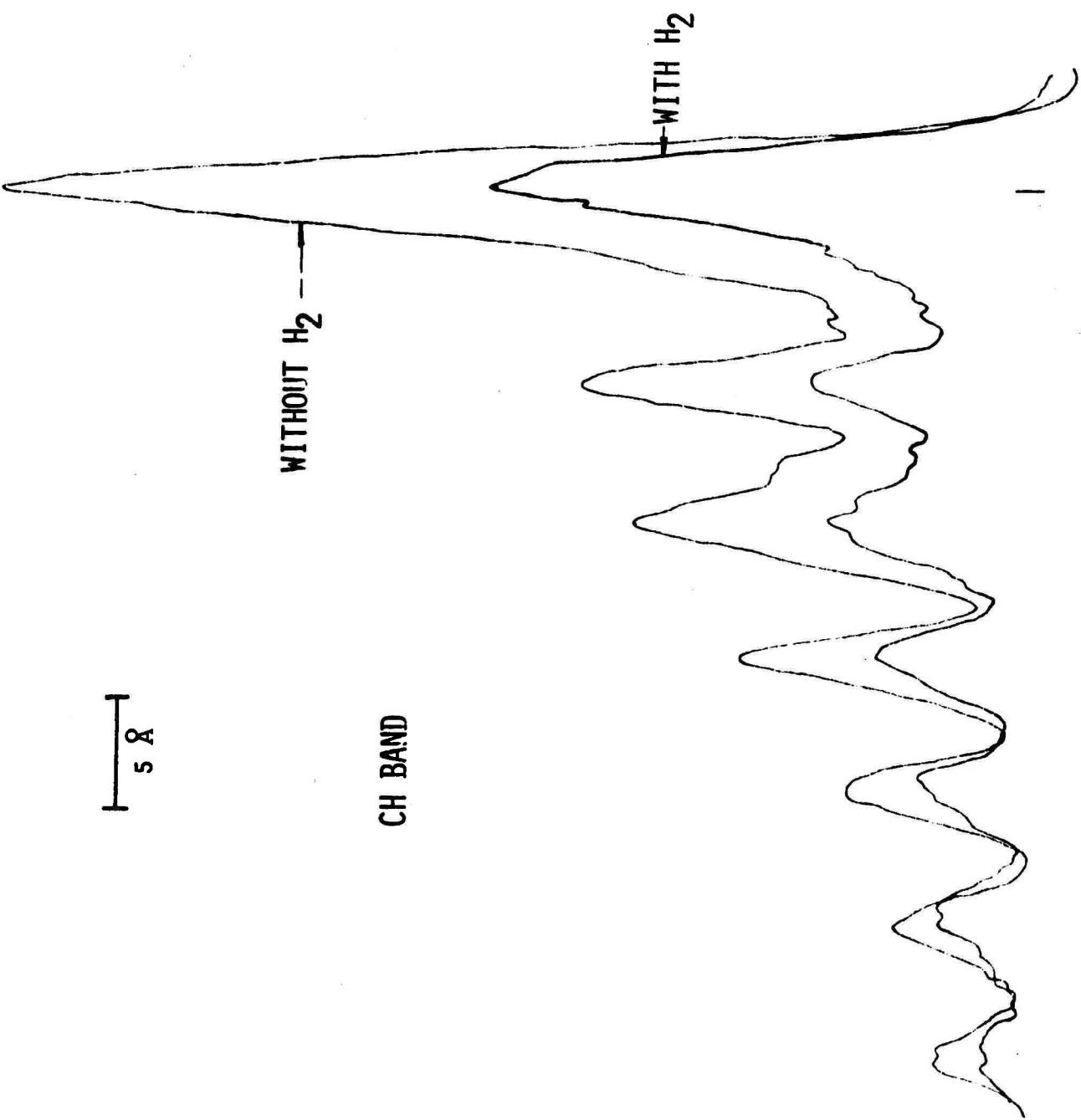


Figure 5

C₂ SWAN BAND

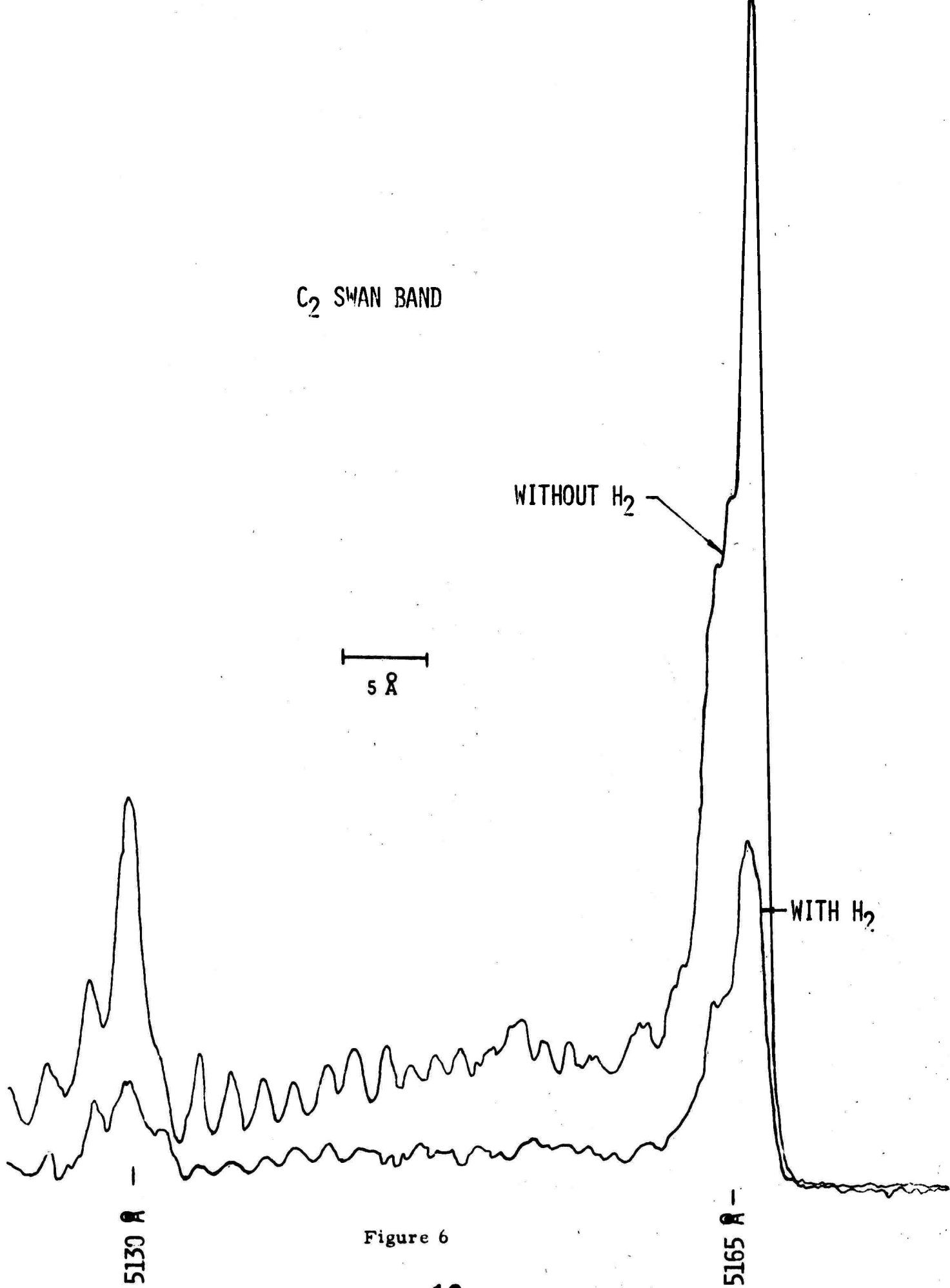


Figure 6

CH BAND

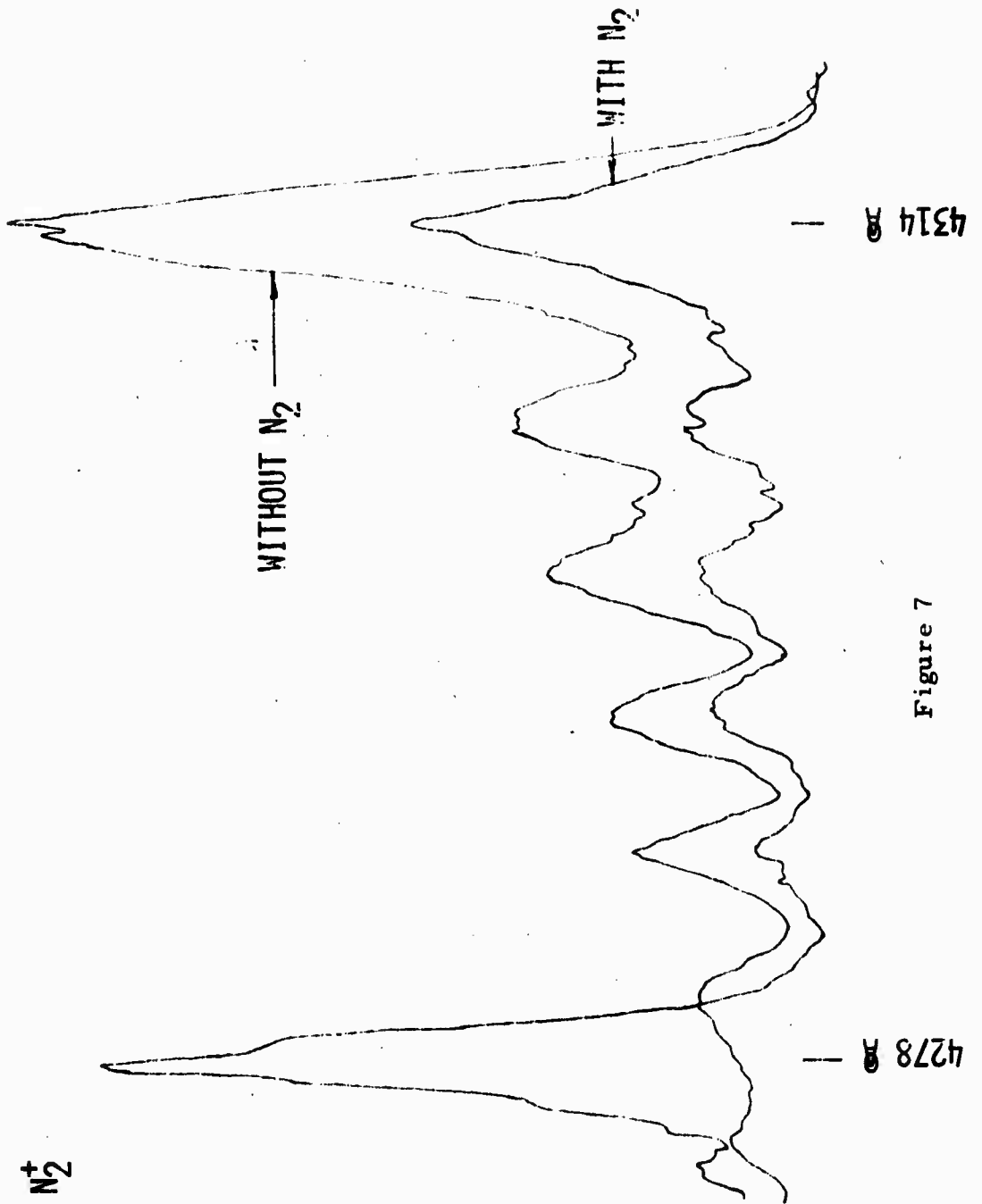


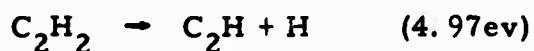
Figure 7

A second possible explanation is that the addition of H_2 lowers the electron "temperature" in the discharge resulting in fewer acetylene molecules being dissociated and, in addition, creates more optimum conditions for generating a population inversion in the gas.

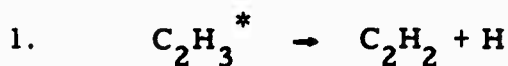
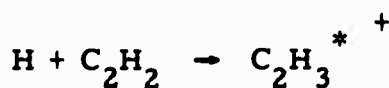
Finally, the possibility exists that the hydrogen prevents the dissociation of acetylene, thus keeping the active specie in the lasing volume for greater periods of time. The energy required to break the triple bond between the carbon atoms is 9.95ev, but once this bond is broken, it is virtually impossible to reform it and these molecules are permanently lost from the lasing process.



In addition, a hydrogen atom can be removed with an energy of 4.97ev leaving the highly reactive C_2H radical.



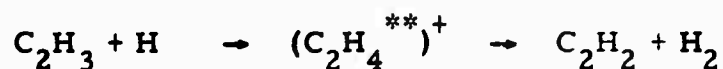
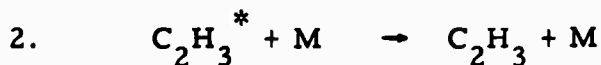
With excess H_2 present in the plasma tube, however, atomic hydrogen is produced (dissociation energy = 4.47ev) and can lead simply to a recombination of atoms without consumption of acetylene. At least two possible reactions exist and start with the hydrogenation of acetylene²:



The * indicates that this molecule is vibrationally excited beyond the dissociation limit. Species without the * may still possess vibrational energy but below the dissociation limit.

Thus, acetylene is reformed in the discharge rather than being split into dissociative products.

The excited $C_2H_3^*$ can also collide with a third molecule reducing the excess energy and then interact with atomic hydrogen to reproduce acetylene and molecular hydrogen.



On the other hand, replacement of hydrogen by nitrogen can lead to the consumption of acetylene by several reactions³. The mechanisms given below are typical.

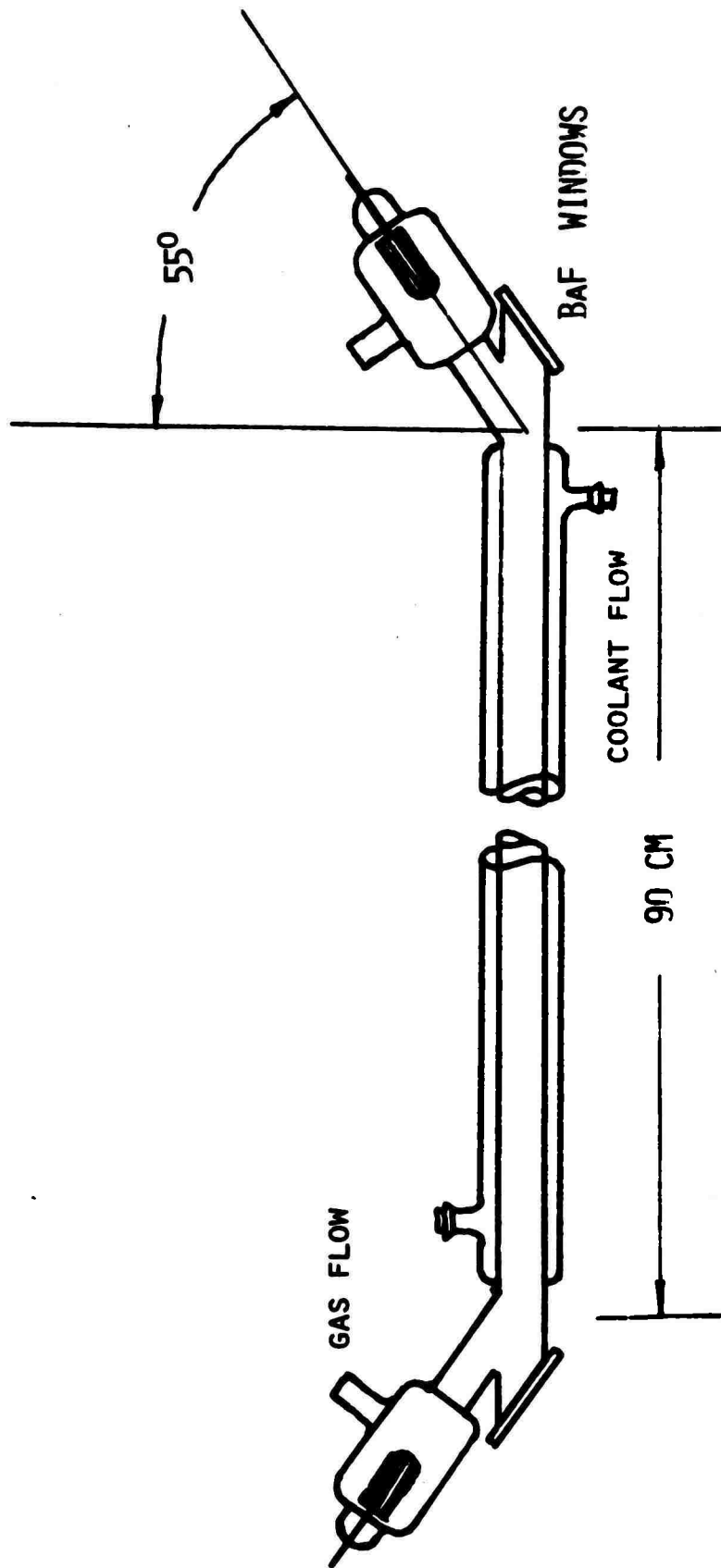


Such destruction of acetylene is consistent with the observation that the substitution of nitrogen for hydrogen is detrimental to the laser performance.

$+(C_2H_4^{**})$ is formed with 63 kcal/mole excess energy above its dissociation energy to form $C_2H_2 + H_2$.²

PLANS FOR FOURTH QUARTER

The plasma tube shown in Figure 8 has been constructed and is about to be placed in operation. The regions of unexcited gas in the optical path have been kept to a minimum, and it is hoped that this will reduce the self absorption on the 3.78 micron transition. High reflectivity mirrors at this wavelength will be installed and we will attempt to get laser action on this transition. Other gases will be tested as time permits.



PLASMA TUBE DESIGNED TO REDUCE UNEXCITED GAS

Figure 8

REFERENCES

1. F. L. Keller, "An Evaluation of the Harmonic and Anharmonic Constants of C_2H_2 from a Reinvestigation of its Infrared Spectrum", PhD Thesis, University of Tennessee (1956).
2. G. G. Volpe and F. Zocchi, *J. Chem. Phys.*, 44, 4010 (1967).
3. D. R. Safrany and W. Jaster, *J. Phys. Chem.*, 72, 3305 (1968).