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(6) OPTICAL PUMPING OF MOLECULAR GASES.

(9) ANNUAL REPORT, 1 Apr 75-31 Mar 76,

for the period

April 1, 1975 to March 31, 1976

(14) G. L. Report No. 2562

(11) April 1976

(12) 38p.

Contract

(15) F44620-74-C-0039

(16) AF-9751 (17) 975102

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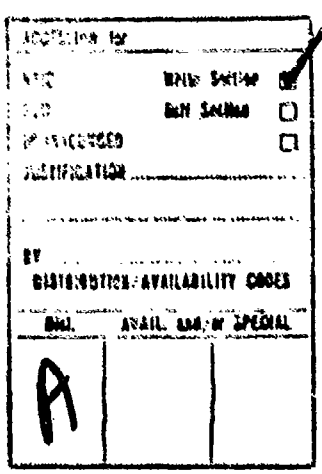
OPTICAL PUMPING OF MOLECULAR GASES

ABSTRACT

During the past year we have completed optical pumping studies of Na_2^{Γ} and have initiated optical pumping studies of CO and Hg_2^{Γ}. Optical pumping of Na_2^{Γ} shows that it is potentially an interesting quasi tunable, high gain dimer laser in the .52 -.81 μm spectra range. Our results show that Na_2^{Γ} could possibly be pumped by a flashlamp or an electrical discharge. Measurements on optically pumping Hg_2^{Γ} show that 2mJ of .265 μm radiation is needed to achieve threshold on the .33 μm band. Experiments are in progress to achieve lasing in Hg_2.



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I. INTRODUCTION

Optical pumping to achieve population inversion and laser action allows detailed studies of potential new laser media without the expense and difficulties involved in discharge pumping. During the past year we have optically pumped I_2 , CO , Na_2 and Hg_2 . The pumping studies of I_2 are complete. We are now using our results to construct an I_2 stabilized Nd:YAG laser by frequency locking the double Nd:YAG laser to $^{127}I_2$ and $^{129}I_2$ absorption lines. Preliminary results of this unique frequency stabilization method have been published.[1]

We are presently using our high energy Nd:YAG oscillator amplifier source pumped tunable $LiNbO_3$ parametric oscillator to optically pump CO . The progress to date is reviewed in Section II. As a result of this work we have gained new insights into population inversion and potential laser action in high pressure CO . In addition, we have invented a new pumping scheme based on V-V transfer from stimulated Raman excited molecular gases. A preliminary experiment in H_2 with CO_2 added shows that the V-V coupling is strong and that laser action should be possible. This pumping scheme does not require a tunable laser source and is therefore a more general method of optically pumping high pressure gases.

We complete a series of optically pumped Hg_2 experiments during the fall of 1975. Our results are discussed in Section III. Based on fluorescence measurements we expect to achieve laser action in Hg_2 with $\sim$4mJ of .2660 μm input pump radiation. Very recently we have begun to pump Hg_2 using a 200mJ Nd:YAG laser pump with an unstable resonator cavity.

With this system we have up to 10mJ of .266 μm for pumping Hg_2 . In our initial experiments we have seen laser action in the Hg atomic system due to $Hg_2^* - Hg_2^*$ collisional excitation of higher lying atomic states.

We have completed our studies of Na_2 by optically pumping. The results are due to be published in the April Journal of Applied Physics. The paper is reproduced as Appendix I. In Section IV we summarize the important results of our optical pumping studies.

II. OPTICALLY PUMPED CO

During the past year theoretical and experimental work has been carried out toward an understanding of the optical pumping of molecular species, in particular CO . As a first step, CO has been vibrationally excited by absorption of radiation at its first overtone band. Next, our study of loss mechanisms for the 2 - 1 vibrational transition revealed more general mechanisms for the redistribution of vibrational energy. That is, emission and reabsorption on overlapping bands in addition to vibration-vibration collisional energy transfer can possibly excite some molecules to a high vibrational state. This prospect will be explored in future work.

Optical Pumping Theory

The overtone pump process considered in this work is diagrammed in Fig. 1. The following assumptions are made:

1. No laser emission during the pump pulse of duration t_p ;
2. Populations of rotational levels in a given vibrational band

are in thermal equilibrium with translational, but not vibrational, degrees of freedom;

3. The pump is not strong enough to saturate one rotation-vibration line before the energy can be collisionally distributed throughout the band.

Let W_{ij} be the stimulated transition probability/time between states i and j, n_{vJ} be the population of a vibration-rotation level, and N_i be the total population of a vibrational state. The rate equations for the overtone pumping system are then,

$$\frac{dN_2}{dt} = W_{0\beta}\left(n_{0\alpha} - \frac{g_\alpha}{g_\beta} n_{2\beta}\right) + W_{J''J'}\left(n_{1J''} - \frac{g_{J''}}{g_{J'}} n_{2J'}\right) \qquad (1)$$

$$\frac{dN_1}{dt} = - W_{J''J}\left(n_{1J''} - \frac{g_{J''}}{g_{J'}} n_{2J'}\right) \qquad (2)$$

$$N = N_0 + N_1 + N_2 \qquad (3)$$

where $g_{J'}W_{J'J''} = g_{J''}W_{J''J}$ by detailed balance, and $g_J = 2\ell + 1$. The rotational states are in thermal equilibrium, so

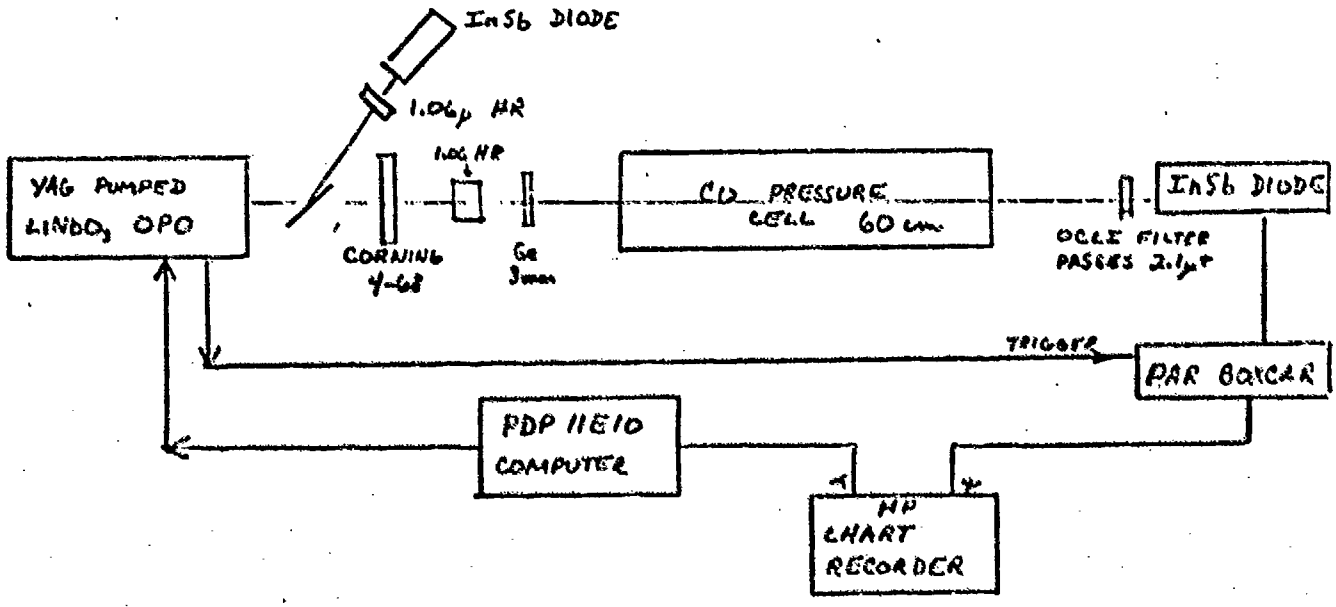

FIG. 1— CO Absorption Measurement Setup

$$n_{vJ} = N_v \left\{ \frac{(2J+1)\exp\left(-\frac{hcB_v}{kT}J(J+1)\right)}{(2J+1)\exp\left[\frac{hcB_v}{kT}(J+1)\right]} \right\}$$

$$\approx N_v g_J \frac{B_v hc}{kT} \exp\left[-\frac{hcB_v}{kT}J(J+1)\right] \equiv N_v X_{v,J} \qquad (4)$$

For CO at $300^{\circ}K$, $X_{\xi=7} = .1$ at the center of the band. Solving for N_2 yields,

$$N_2 = \frac{N\left\{1 - \exp\left[-W_{\alpha\beta}\, t \left(X_2 + \frac{g_\alpha}{g_\beta} X_\beta\right)\right]\right\}}{1 + \frac{g_\alpha}{X_\alpha}\frac{X_\beta}{g_\beta}} \qquad (5)$$

Since $Wt_p = It_p \sigma_{20}/h\nu$, where I is pump intensity and σ_{20} is the overtone cross section for the transition at ν, the saturation pump energy per unit area $\mathcal{E}_p$ is found to be

$$\mathcal{E}_p = \frac{h\nu_{20}}{\sigma_{20}} \frac{1}{X_\beta\left(1 + \frac{g_\beta}{g_\alpha}\frac{X_\alpha}{X_\beta}\right)} \qquad (6)$$

At room temperature the $v = 1$ band has almost no population so that virtually all optically pumped molecules have the inversion,

$$\Delta n = N_2 \chi_{J'} = \begin{cases} \dfrac{N\chi_{J'}\chi_\alpha \sigma_{20}}{h\nu_{20}} \dfrac{g_\beta}{g_\alpha} \mathcal{E}_p & \text{small pumping} \\ \dfrac{N\chi_{J'}}{1 + \dfrac{g_\alpha}{\chi_\alpha}\dfrac{\chi_\beta}{g_\beta}} & \text{saturation pumped} \end{cases} \tag{7}$$

Gain for a transition in the 2 - 1 band of cross section σ_{21} is given by,

$$\gamma = \sigma_{21} \Delta n$$

So

$$\chi = \begin{cases} \dfrac{N\chi_{J'}\chi_\alpha \sigma_{20}\sigma_{21}}{h\nu_{20}} \dfrac{g_\beta}{g_\alpha} \mathcal{E}_p & \text{small pumping} \\ \dfrac{N\chi_{J'}\sigma_{21}}{1 + \dfrac{g_\alpha}{\chi_\alpha}\dfrac{\chi_\beta}{g_\beta}} & \text{saturation} \end{cases} \tag{8}$$

For CO , integrated cross sections in the Doppler broadened regime are,

$$\sigma_{20} = 3.4 \times 10^{-19} \text{ cm}^2 \quad ^2$$

$$\sigma_{21} = 2.1 \times 10^{-15} \text{ cm}^2 \quad ^3$$

In the pressure broadened regime these become for a single vibrational rotational line near midband,

$$\chi = \begin{cases} \sigma_{20}(7) = 2 \times 10^{-21} \text{ cm}^2/\text{P(atm)} \equiv \sigma'_{20}(7)/\text{P(atm)} \\ \sigma_{21}(7) = 1.4 \times 10^{-17} \text{ cm}^2/\text{P(atm)} \equiv \sigma'_{21}(7)/\text{P(atm)} \end{cases}$$

The gain is then

$$(1.34)\, \mathcal{E}_p(\text{J/cm}^2) \quad 1/\text{P(atm)} \quad \text{cm}^{-1} \qquad \text{small pump,}$$

$$16.7 \text{ cm}^{-1}$$

Saturation pumping occurs for $\mathcal{E}_p \geq 2.5\ [\text{P(atm)}]\ \text{J/cm}^2$.

For such high gains, superfluorescent emission should be possible. If bleaching is negligible, the pump pulse has the spatial decay

$$\mathcal{E}_p(z) = \mathcal{E}_p(0) e^{-\alpha_{20} z} \qquad (9)$$

Also let $\gamma(z) = \mathcal{E}_p(0)e^{-\alpha_{20}z}$, where $\beta = (1.34)/P(atm)$. Then

$$\ln\left(\frac{P_{em}}{P_o}\right) = \int_o^{\ell} \gamma(x)\,dt \approx 4$$

where ℓ is the gain length required for the pulse to build up. Solving for ℓ yields,

$$\ell = \frac{-kT}{\sigma_{20}P} \ln\left\{1 - \ln\left(\frac{P_{em}}{P_o}\right)\left(\frac{h\nu_{20}g_\alpha}{\sigma'_{21}\chi_J,\chi_\alpha g_\beta}\right)\left[\frac{P(atm)}{\mathcal{E}(0)}\right]\right\} \qquad (10)$$

Experimental Work

The large required pump intensities have made desirable the use of the high energy Nd:YAG pumped $LiNbO_3$ optical parametric oscillator (OPO) currently under development at Stanford. The tuning of this device from 1.5 - 4.5 μm is under the control of a PDP11E/0 computer with a CAMAC interface to a pair of stepping motors which vary the angles of the OPO crystal and grating. The output frequency of the device is scannable over its entire tuning range at a rate which is interactively variable in real time by the experimeter. For higher resolution, a 1 mm fused silica etalon is used in the OPO cavity and is driven by the computer to give a linear tuning response.

In order to ascertain the optimum pumping frequency and to demonstrate our device's scanning capability, an absorption scan was taken of

the CO first overtone band near 2.3 μm . The experimental setup is shown in Fig. 2 and the resulting chart recorder trace in Fig. 3. Note the rotational line structure in the P branch. Structure in the R branch is not observable due to the 2 cm^{-1} bandwidth of the OPO.

To obtain a measurement of the cross section for a single rotational line, the etalon was placed in the OPO and a scan over 4 cm^{-1} was performed over the R(7) and R(8) lines, as shown in Fig. 4. At 1 atm the measured cross section is 2.2×10^{-21} cm^2 , in agreement with the literature. This value has been corrected for the relating linewidth of the OPO (~ .4 cm^{-1}) and CO(.14 cm^{-1} at 1 atm)[4] .

Next an attempt to observe superfluorescent emission on the 2 - 1 band near 4.6 μm was made. From Eq. (10) it is seen that $\mathcal{E}_p/P$ has a threshold which is calculated for CO to be

$$\frac{\mathcal{E}_p(0)}{P(\text{atm})} = 2.3 \text{ J/cm}^2 - \text{atm} \quad .$$

This value was exceeded in many attempts in which ℓ was 3 - 4 times the beam confocal parameter, or focal depth of field, but no stimulated emission was observed.

Discussion

The negative result for emission spurred an investigation of additional loss mechanisms in CO . The 2 - 3R and 0-1R bands slightly overlap the 2-1R

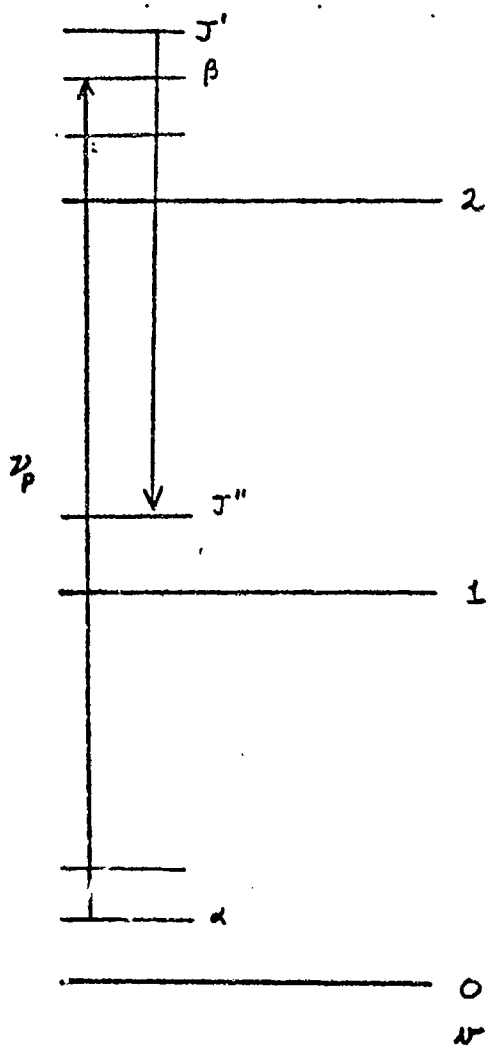

FIG. 2-- Overtone Pumping Level Scheme

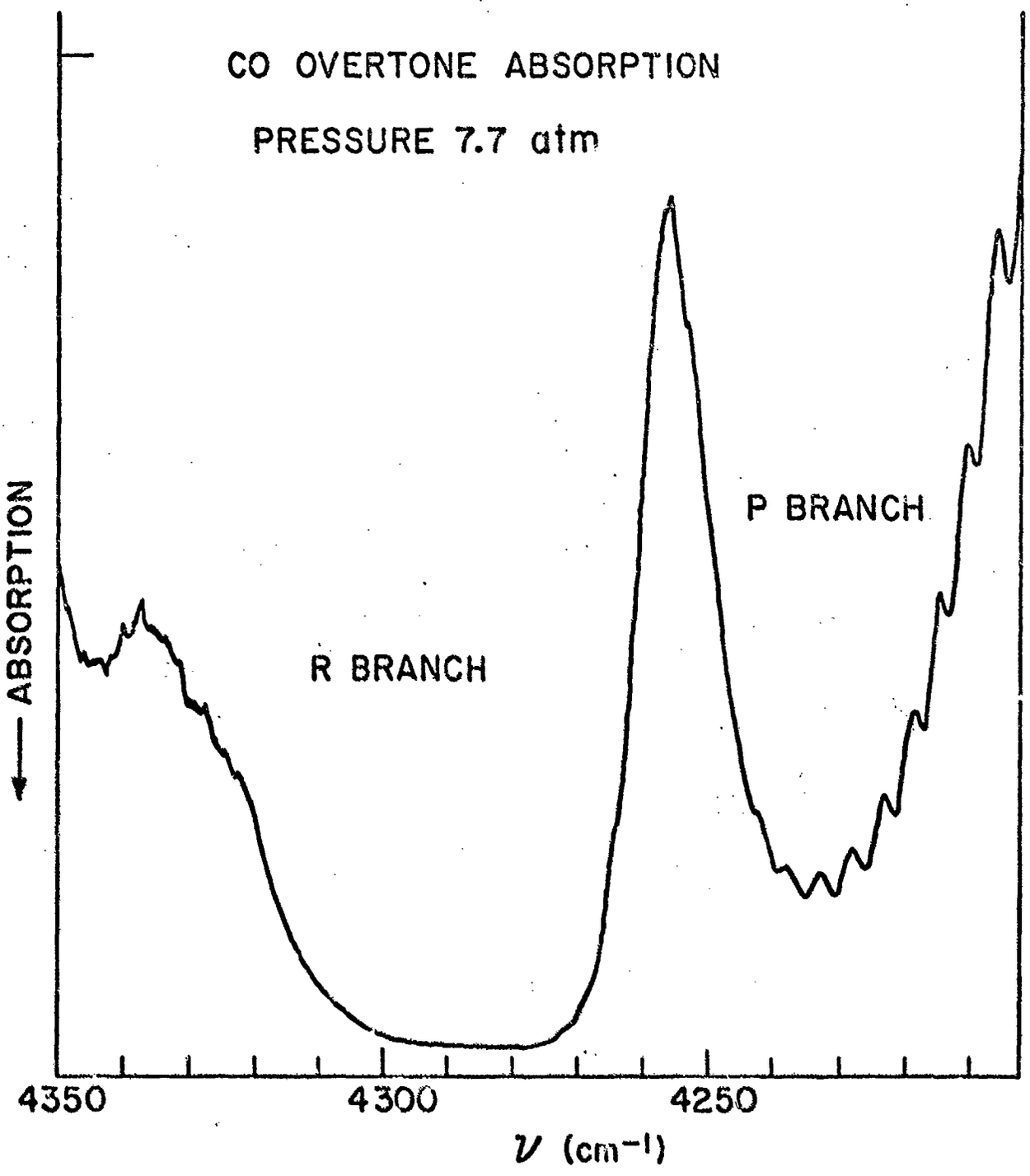

FIG. 3--Overtone absorption spectrum of CO at 7.7 atm pressure taken with the computer controlled tunable source.

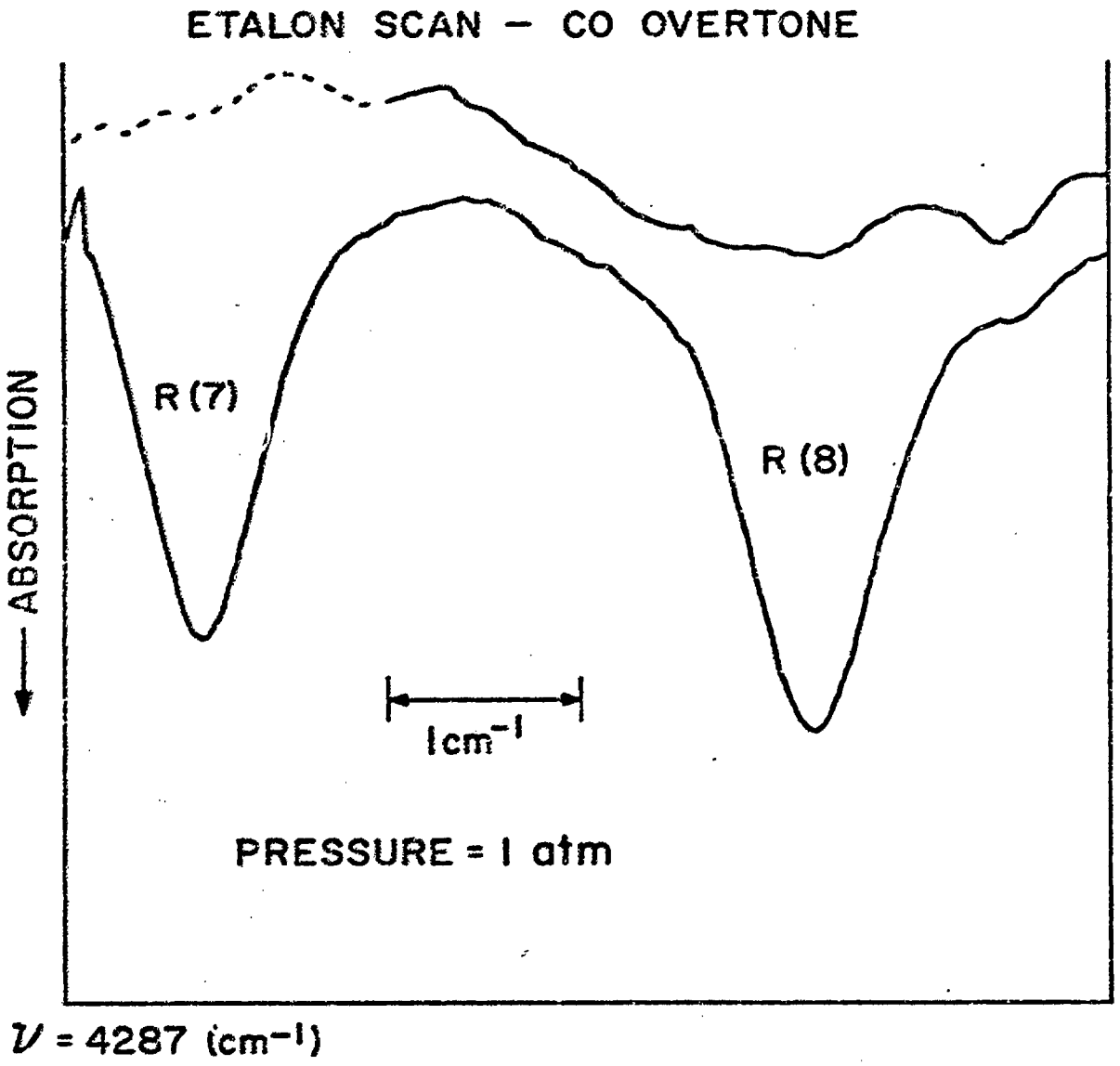

FIG. 4--Resolved rotational lines of CO using the etalon narrowed tuneable source.

and it was hoped that emission might be observed for the 2 - 1 R(10 - 15) lines, but possibly the large number of ground state molecules makes even a weak overlapping line strong when compared to the 2 - 1 transition. In this case stimulated emission may not be possible.

However, the prospect of overlapping bands for the 2 - 3 transition makes possible a radiative mechanism for redistribution of optical pumping absorbed energy to high states of vibrational excitation. A CO molecule so excited may then transfer its energy to another specie which has a state closely matched in energy with the excited CO state. Such vibration-vibration (V-V) or electronic-vibration (E-V) transfer mechanisms can be very fast for small energy defects. A V-V transfer effect also will play an important role in the CO energy redistribution, particularly since V-V upward transitions are more probable than downward ones.[5]

To pursue this, we plan to measure the fluorescence spectrum of optically excited CO in order to determine the extent to which CO may become collisionally or radiatively excited. These experimental results will then be compared with a computer generated solution to the coupled vibrational rate equations which was solved earlier this year.

III. OPTICALLY PUMPED Hg_2

An attempt to observe stimulated emission on the 335 nm ultraviolet band of Hg_2 excimer by laser pumping was made in a series of experiments using a new mercury cell. Although no stimulated emission has been observed yet, the measurements on the UV and the visible fluorescence bands indicate

that the stimulated emission is possible for both transitions with increased laser pumping energy at 266 nm. The experimental results and the future plans are summarized below.

Mercury Cell

Previous experience has shown that extreme care in cleanliness must be taken in order to fill a cell with mercury. The present 30 cm-long quartz cell with Brewster angle windows and a side arm reservoir for mercury was first chemically cleaned and then baked in an oven at 450°C for several hours while the cell was evacuated to less than 10^{-7} torr. The bake-out procedure was necessary in order to drive out adsorbed gases on the cell wall. The cell was filled with mercury by slow distillation and sealed off.

As a preliminary check for purity, the cell was heated to about 300°C and was probed with a HeCd laser at 441.6 nm. No visible fluorescence was observed, indicating that there were no appreciable contaminants that can absorb 441.6 nm.

Pump Energy Absorption by Hg_2

Laser induced fluorescence of Hg_2 was studied with a pump wavelength of 266 nm generated by quadrupling Nd:YAG 1.064 μm in two doubling crystals CDA and ADP. Absorption of 266 nm was measured at various atomic mercury vapor densities ranging from 10^{18} to $10^{19}/cm^3$. The Hg density was temperature controlled in a side-arm mercury reservoir while the cell was

kept nearly at a constant temperature of about $400^{\circ}C$.

The attenuation parameter k may be expressed in the form $I(\ell)/I(0) = \exp[-kN^2\ell]$. The measured value of k was $(6.5 \pm 2.2) \times 10^{-40}$ cm^5 . This value is in agreement with a value obtained by the group at National Bureau of Standards (7.27×10^{-40} cm^5) .

Hg_2 Fluorescence Data

The quantitative measurements of Hg_2 fluorescence as mostly limited to the visible band data primarily due to the fact that the operating conditions of the mercury cell significantly favored population of excited molecules in the state associated with the visible band. The UV band fluorescence was definitely observed, however, its intensity was an order of magnitude smaller than the visible band. The UV detection was made difficult and less reliable because of the residual visible fluorescence band which overlaps with the UV band near 350 ~ 370 nm range.

The Hg_2 band fluorescence measurements consisted of fluorescence decay rate and relative intensity as a function of atomic mercury density at a fixed cell temperature of about $400^{\circ}C$. The measured values of the visible fluorescence decay constant τ were 37 μsec, 28 μsec, and 24 μsec at densities of $3.3 \times 10^{18}/cm^3$, $4.4 \times 10^{18}/cm^3$, and $5.6 \times 10^{18}/cm^3$, respectively. An estimated error of measurement is about $\pm$ 2 μsec, but a larger uncertainty may be attributed to the values of number density. The UV fluorescence decay showed τ of about 100 nsec or less, where the resolution was limited by oscilloscope response.

The fluorescence band intensity versus number density data are shown in Figs. 5 and 6 for the visible and the UV bands, respectively. In an attempt to understand the shape of the respective intensity plots, empirical formulas for the excited state population distribution were derived using the relationship $\log[I(\text{visible band})/I(\text{UV band})] = N/N_T$ from NBS data. The population in each state can be expressed as follows:

$$n_0(\text{visible}) = n^* \left[1 + \frac{N_T}{N}\right]$$

$$n_1(\text{UV}) = \left[1 + \frac{N}{N_T}\right]$$

$$n^* = (E_{pump}/\hbar\omega_{pump}) \left[1 - e^{-kN^2 l}\right] ,$$

where N is the mercury density, n^* is the number of pump photons absorbed, and N_T is the number density at which $n_1 = n_0$. The formulas are valid for $T_{vapor} > 300^0C$ and $N > 5 \times 10^{17}/cm^3$.

The graph of n_0 and n_1 versus N are respectively shown in Figs. 7 and 8 with a suitable n^* to match the intensity scale of Figs. 1 and 2. The qualitative correlation for the visible band is very good while that of the UV band shows a general agreement in shape. The validity of the form of the empirical formulas is well supported by the agreement with experimental observation.

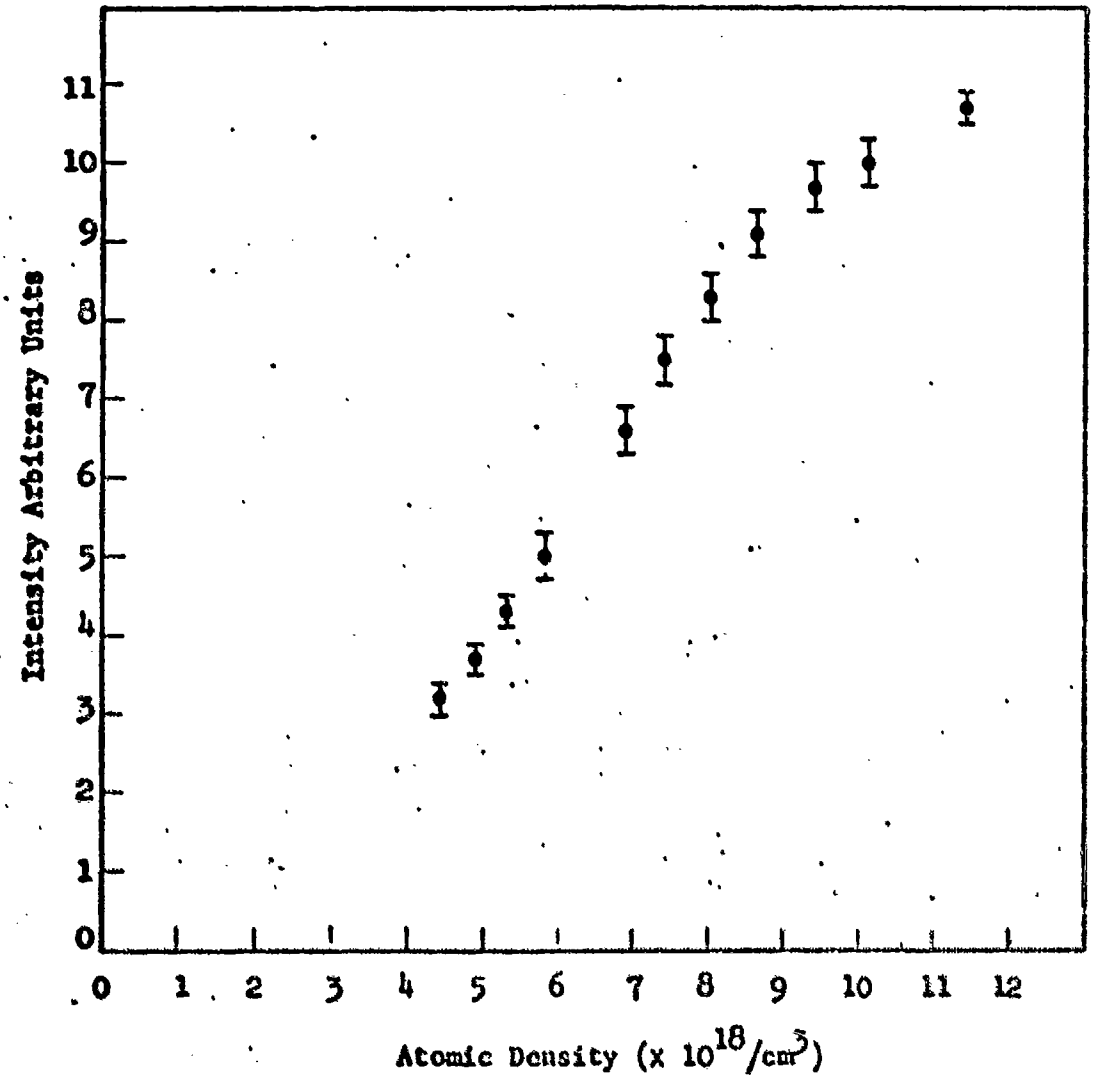

FIG. 5--4850 Å visible band fluorescence intensity vs atomic density T ≈ 410°C .

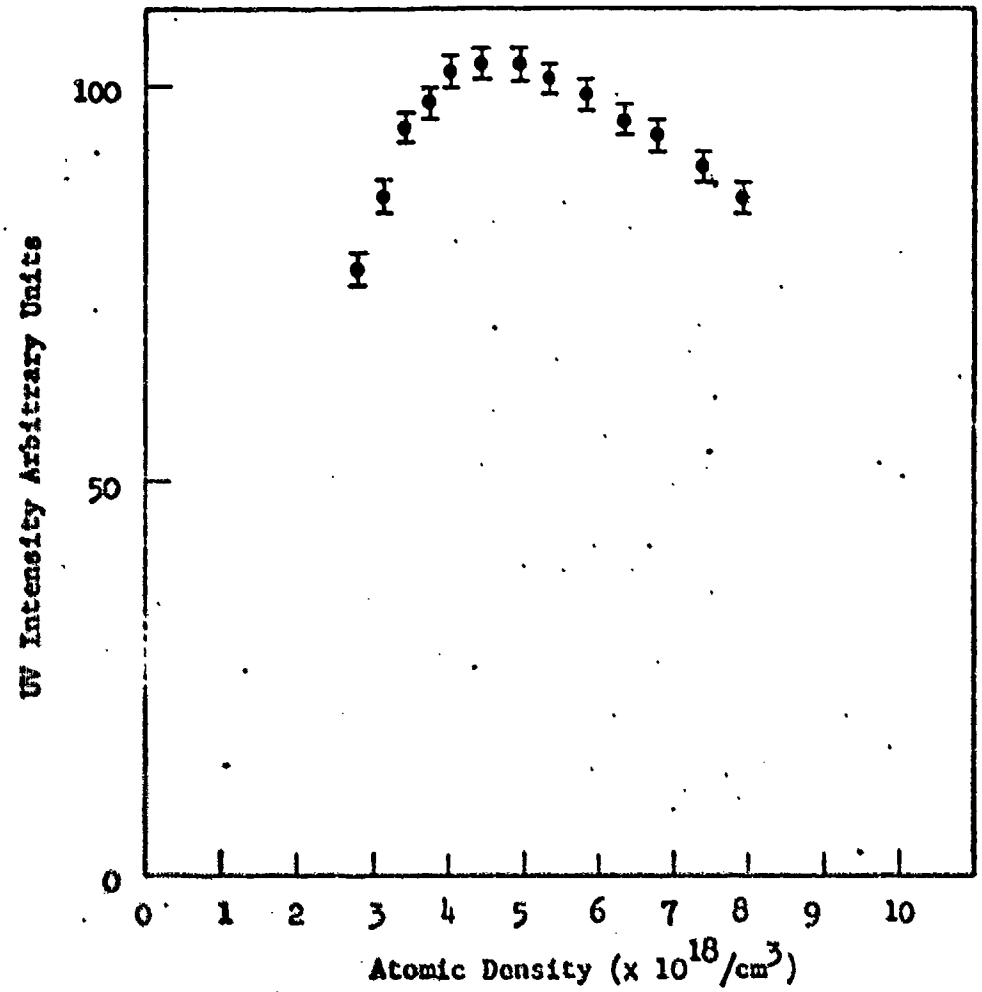

FIG. 6--UV band fluorescence intensity vs Hg density ($T_{cell} \approx 380^{\circ}C$).

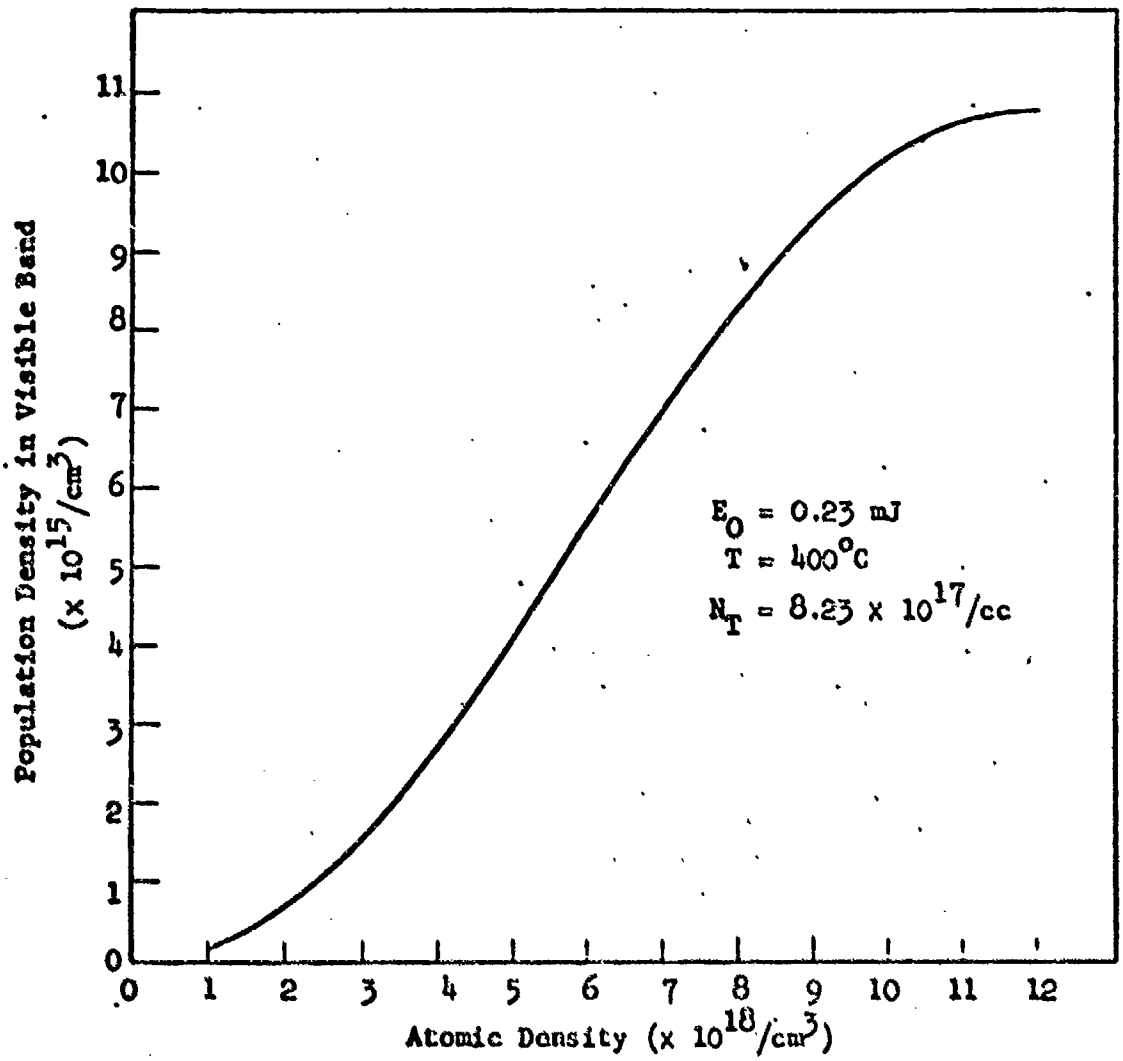

FIG. 7--Population density n_0 in the visible band vs atomic density at a cell temperature of 400°C. The incident .226 μm energy is .234 mJ focused to a 20 cm confocal distance.

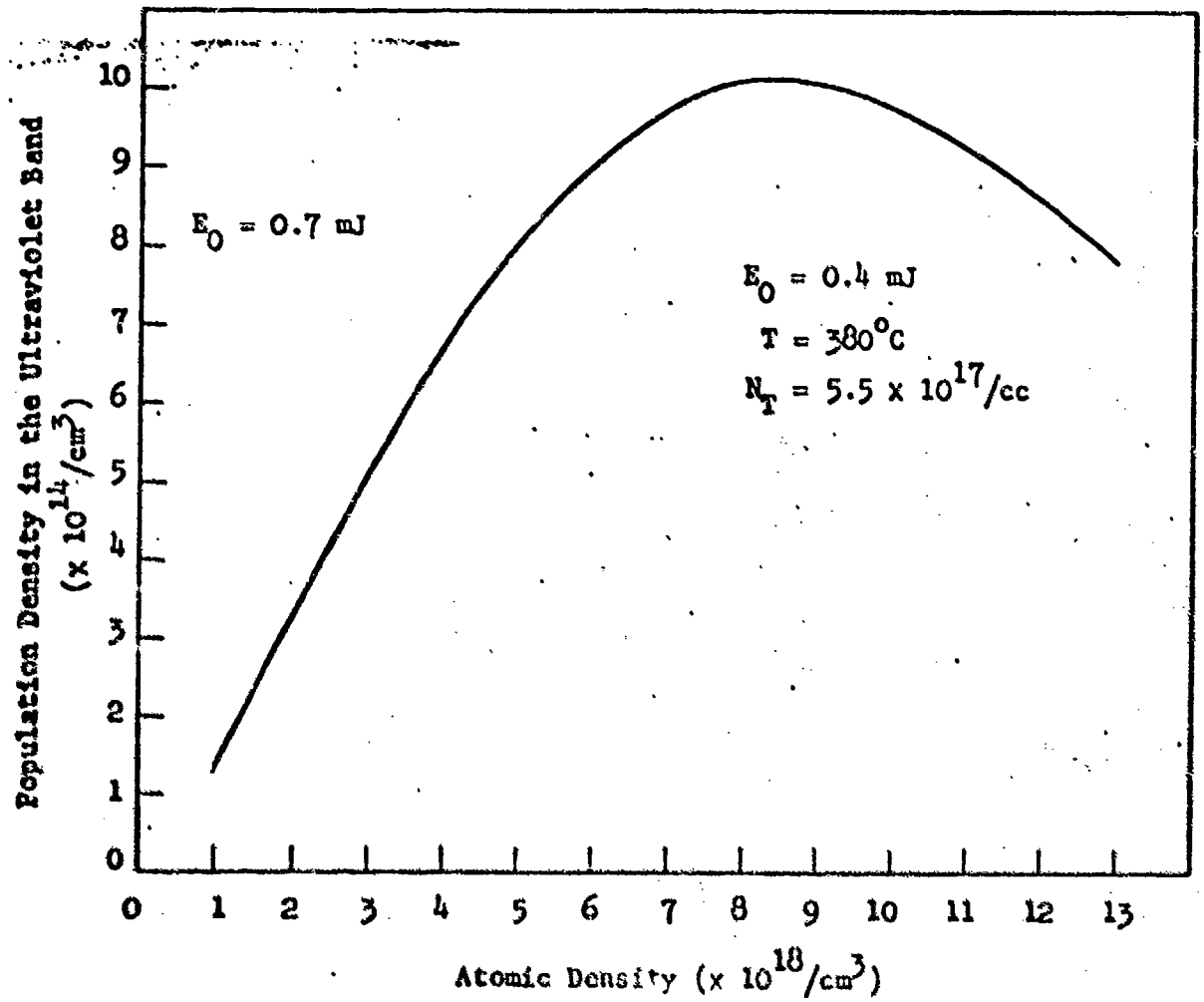

FIG. 8--Population density n_1 in the ultraviolet band vs atomic density at a cell temperature of 380°C. The incident 0.266 μm energy is 0.40 mJ focused into a 30 cm confocal distance.

The empirical formulas for n_0 and n_1 can now be used to predict excited state population at given values of pump energy and number density. A plot of n_0 and n_1 vs N at a fixed pumping energy of 1 mJ at 266 nm is shown in Fig. 9 (assuming all of absorbed energy is distributed into n_0 and n_1) . The plot indicates that n_0(visible) dominates over n_1(UV) at a vapor temperature of $\sim 400^{\circ}C$. First, an order of magnitude difference in the populations explains the smaller UV fluorescence intensity in our experiments. Second, n_1(UV) shows a peak value near $N = 8 \times 10^{18}/cm^3$ which sets an optimum condition for n_1 . The position of the peak is dependent on temperature, but it is not significantly affected in the range of temperatures in our experiment.

Laser Oscillation Threshold

The threshold conditions for laser oscillation can be derived from fluorescence life time and inversion density. Based on experimental data, stimulated emission cross-section of $\sim 1 \times 10^{-20}$ cm^2 and $\sim 3 \times 10^{-18}$ cm^2 are calculated for the visible and the UV bands, respectively. Assuming an active region of 3×10^{-2} cm^3 in a 30 cm cell, the minimum pump energy required to achieve oscillation is approximately 1.7 mJ and 2.5 mJ at $N = 5 \times 10^{18}/cm^3$ for the visible and the UV bands, respectively.

The energy of 266 nm laser available at the time of experiment was less than 1 mJ. This accounts for the absence of stimulated emission in our experiment.

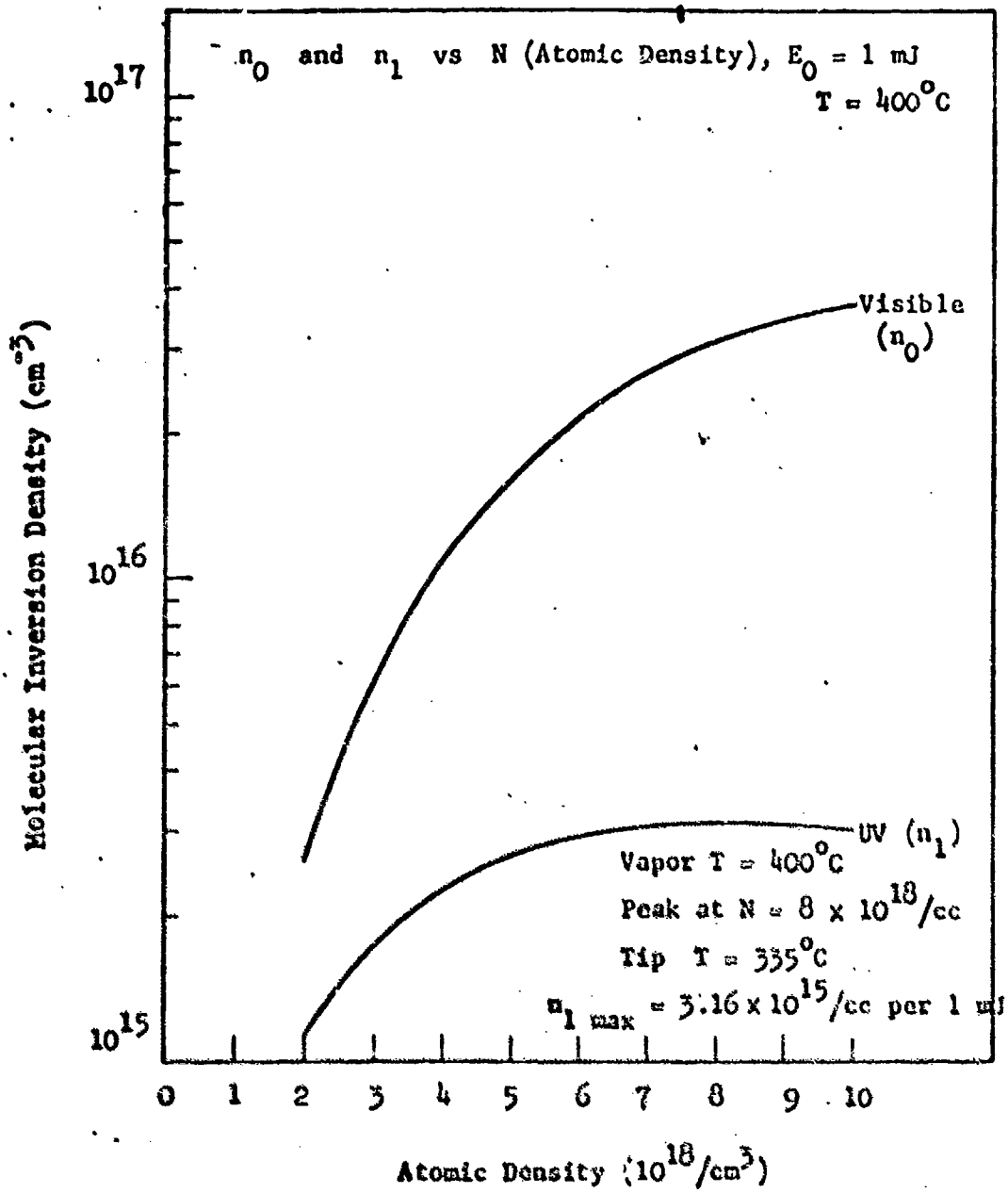

FIG. 9--Graph of n_0 and n_1 as a function of N at fixed vapor temp. T = 400°C.

Assumed Parameters: E_{laser} = 1 mJ , V = 10^{-3} cm^2 x 30 cm (l)

$kl = 2.18 \times 10^{-38}$ cm^6 $N_T = 8.23 \times 10^{17}$/cc (From NBS data)

$$n_0 \doteq 4.5 \times 10^{16} \left[1 - e^{-klN^2}\right]\left[1 + \frac{N_T}{N}\right]^{-1} \qquad n_1 \doteq 4.5 \times 10^{16}\left[1 - e^{-klN^2}\right]\left[1 + \frac{N}{N_T}\right]^{-1}$$

Future Experiments

Presently, a Nd:YAG oscillator system capable of 200 mJ output energy is operating. Angle-phased matched KD^*P and ADP crystals for harmonic generation are expected to deliver more than 10 mJ at 266 nm. The increased pump energy offers a greater probability of success for stimulated emission in Hg_2 excimer system.

IV. OPTICALLY PUMPED Na_2

The details of our optical pumping studies of Na_2 are presented in Appendix I. The study showed that Na_2 is capable of lasing on both A and B bands at low input pump energies. The required optical pumping powers are obtainable from flashlamps so that it should be possible to flashlamp pump Na_2. Furthermore, Na_2 is stable in a discharge and is known to be an efficient fluorescing molecule in a discharge. It is therefore probable that Na_2 can be discharge excited to provide output radiation over the .5- .8 μm spectral range. The related dimer molecules K_2, Rb_2 and Cs_2 should allow the wavelength range to extend to 2 μm.

V. RESEARCH DIRECTIONS FOR NEXT QUARTER

During the next quarter we plan to continue our optical pumping studies of CO and Hg_2. For CO we plan to more closely investigate lower level absorption that may be supressing laser action. We also plan to investigate UV transfer pumping using vibrational energy stored in CO or Raman induced vibrational energy in H_2.

APPENDIX

OPTICALLY PUMPED SUPERFLUORESCENT Na_2 MOLECULAR LASER

by

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Optically pumped superfluorescent Na_2 molecular laser

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(Received 8 October 1975; in final form 22 December 1975)

We have optically pumped an Na_2 molecule at 0.473 and 0.659 μm and obtained superfluorescent laser emission on a series of green and yellow molecular B-X-band transitions and laser oscillation in the near infrared on A-X-band transitions. Pump saturation, laser gain, and laser saturation intensity are reported.

PACS numbers: 42.60.Cz, 32.20.Ft, 32.20.Ky

Optical pumping is a convenient method to quantitatively study new potential lasers prior to attempting the more difficult electrical discharge and flash-lamp pumping methods. By optical pumping one can directly measure pump absorption and saturation intensity, induced laser gain and laser saturation intensity, and the effects of quenching and buffer gas pressure.

We choose to study the Na_2 dimer because of its relatively well-known spectrum,[1] potential high laser gain, and potential for laser emission over the visible to near near-infrared spectral range from 0.54 to 0.81 μm on both the $B\,^1\Pi_u$-$X\,^1\Sigma_g^+$ and $A\,^1\Sigma_u^+$-$X\,^1\Sigma_g^+$ molecular bands.[2,3] We observed superfluorescent laser emission on a series of green and yellow molecular B-X—band transitions and laser oscillation in the near infrared on A-X—band transitions. Na_2 is representative of the other alkali dimer systems which have molecular emission bands extending from the visible into the infrared to beyond 1.2 μm for the case of Cs_2.

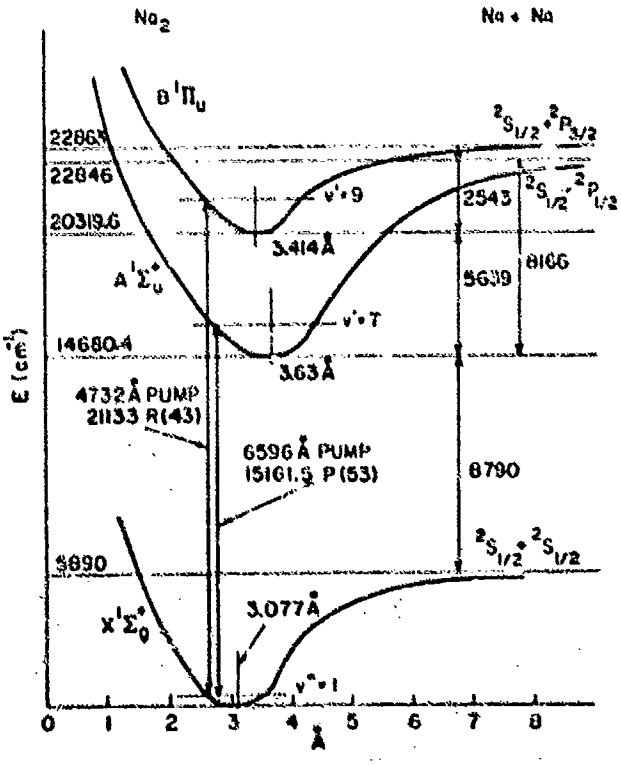

FIG. 1. Energy-level diagram of molecular Na_2 showing 4732- and 6596-Å pump wavelengths used to achieve inversion in the B and A bands.

Figure 1 sketches the energy-level diagram of molecular Na_2. In our experiments we pumped the B band of Na_2 at 4730 Å generated from a doubled Q-switched Nd:YAG laser. We also pumped the Na_2 A band at 6594 Å using a doubled Q-switched Nd:YAG laser operating at 1.32 μm. Finally, we used a Molectron N_2 pumped dye laser operating over the 4500—5000-Å range of coumarin 102 dye to optically pump Na_2. Bright superfluorescent laser emission every 1.5 Å of the pump wavelength spacing was observed over a pump band from 4627 to 4885 Å. These experiments demonstrate the high gain and ease of laser action in Na_2.

The typical experimental apparatus is shown in Fig. 2. For B-band optical pumping, the 4730 Å was generated by a Chromatix model 1000 Q-switched Nd:YAG laser operating at 0.946 μm and internally doubled with a $LiIO_3$ crystal. The bright visible superfluorescent emission was photographically recorded through a 1-m Chromatix grating spectrometer against an atomic-potassium reference spectrum. The 4730-Å radiation was loosely focused through antireflection-coated fused-silica windows of a small stainless-steel heat pipe containing a 6-cm column of metallic sodium vapor in a helium buffer gas. At a vapor-column temperature of 605 °C, corresponding to $p_{Na_2}/p_{Na} = 1.4$ Torr/28.5 Torr, with a buffer gas pressure of 30 Torr, the superfluorescent B-band laser was found to have a pumping threshold energy of 9 μJ. With increasing buffer gas pressure the laser threshold rapidly increased with 110 Torr being recorded as the upper limit for observing sporadic laser action, with a sodium temperature of 605 °C and a maximum pump energy of 225 μJ. Below about 30 Torr of buffer gas, fogging of the cell windows was evident. An average superfluorescent pulse energy of 1.31×10^{-1} μJ was measured, indicating an energy conversion efficiency of 0.067%, which is less than expected from a three-level model for this laser. The calculated saturated gain of 4.07 cm^{-1} per pump transition based on the measured superfluorescent output energy, assuming approximately nine strong pump transitions within the pumping bandwidth, is high, as expected.

The observed Na_2 laser transitions for the 4730-Å pump are shown in Table I. The table shows two sets of transitions which were separated by noting that the second set shows constant differences. The top set predominantly fit B-band Q-branch transitions corresponding to the Q-branch pump lines given in Table II. The second set of lines have tentatively been identified as A-band transitions terminating on $v''=3,4,5$ for J''

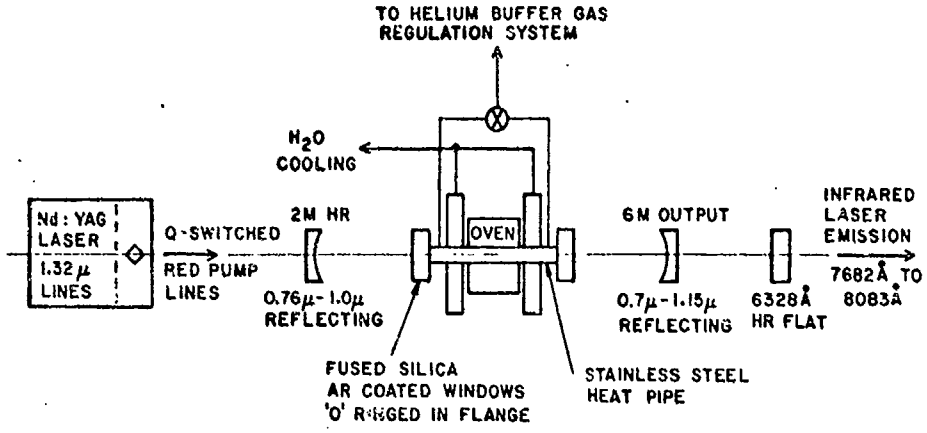

FIG. 3. Experimental layout for optically pumping Na_2 which is contained in a small heat-pipe oven.

$= 6, 7, 8$ of the $X^1\Sigma_g^+$ ground electronic state.[4] If this assignment is correct, then we have observed a B-A—band collision-induced crossover with an energy defect of nearly 1950 cm^{-1}.

To further investigate the possibility of B-A—band crossover the laser output was observed on a fast oscilloscope to determine its time dependence. It was immediately evident that two different pulse lengths occurred. One output pulse consisted of a single 20-nsec spike occurring near the beginning of the 1-μsec 4730-Å pump pulse. The second time behavior consisted of a 100—200-nsec pulse. With the spectrometer tuned near 5300 Å as a broad-band filter, the longer 100—200-nsec pulse was consistently observed. This long pulse behavior is consistent with the fact that superfluorescent A-band emission near 5300 Å following band crossover requires relaxation of approximately 16 vibrational levels in the A electronic state to account for the 1950-cm^{-1} energy defect. It is not understood why such a band crossover occurs or to where the excess energy goes. Further studies must be made to determine the mechanism of the energy transfer.

We attempted to identify the possible pump bands covered by the 2-cm^{-1}-wide 4730-Å line. The wavelength in vacuum is 4731.81 Å or 21133.6 cm^{-1}. Using molecular constants for Na_2 from Demtroder *et al.*,[5] we tentatively identified a least nine B-band transitions within the 4730-Å linewidth. These pump lines and assignments are shown in Table II along with the relative ground-state population of the transitions at 600 °C. At this temperature the absolute density of Na_2 molecules is 1.35 Torr or $N'' = 1.49 \times 10^{16}$ cm^{-3} with a maximum population at $J'' = 44$. Vibrational levels up to $v'' = 5$ have an appreciable population at the 600 °C vapor-column temperature. The fraction of the total ground-electronic-state population in the nine pump transitions is 1.27×10^{-3} from Table II which corresponds to an average population of 2.10×10^{13} cm^{-3} for each pump transition.

In an additional experiment to determine the possibility of eventually flash-lamp pumping the Na_2 molecule, we pumped the B band of the molecule with a tunable dye laser. Using a Molectron N_2 pumped dye laser which tuned over the 4600—4900-Å range of coumarin 102 dye with a 6-nsec pulse width at 50—150 kW peak power, we observed bright yellow-green superfluorescent laser emission, at pump spacings of approximately 1.5 Å over a range from 4627 to 4885 Å. Over 170 strong pump bands were observed over this wavelength range. The dye-laser pump linewidth was approximately 2 cm^{-1}, so with an average spacing of about 0.2 cm^{-1} between strong pump transitions, we estimate that about 10 pump lines occurred under the dye-laser bandwidth. These experiments were run at a sodium-vapor-column temperature of 535 °C, corresponding to an Na_2 partial pressure of 0.35 Torr and Na partial pressure of 7.5

TABLE I. B-band laser lines observed with a 4730-Å pump.[a] λ pump (air) = 4730.5 Å; λ pump (vacuum) = 4731.81 Å/21133.6 cm^{-1}.

λ air	λ vac	ν vac (cm^{-1})
5491.58	5493.1	18204.7
5446.94	5448.45	18353.8
5413.11	5414.61	18468.5
5402.44	5403.94	18505.0
5378.14	5379.63	18588.6
5369.02	5370.51	18620.2
5349.30	5350.78	18688.9
5263.33	5264.79	18994.1
5366.35	5367.84	18660.3
5384.97	5386.46	18595.1
5342.83	5344.31	18711 5
5341.50	5342.98	18716.1
5299.52	5300.99	18864.4
5298.16	5299.63	18869 2

[a]Wavelength accuracy to within ± 0.1 Å.

TABLE II. Probable B-band pump transitions for 4730-Å pump. λ pump (air) = 4730.5 Å; λ pump (vacuum) = 4731.81 Å/21133.6 cm^{-1}.

Vacuum wave numbers	v'', J''	v', J'	Branch	Relative population at 600 °C
21132.6	1,37	9,36	$P(37)$	2.374×10^{-1}
21132.8	6,51	17,52	$R(51)$	6.389×10^{-4}
21133.0	1,43	9,11	$R(43)$	2.432×10^{-3}
21133.2	5,39	15,39	$Q(39)$	8.418×10^{-4}
21133.4	2,52	11,51	$P(52)$	1.811×10^{-3}
21133.6	4,53	14,53	$Q(53)$	1.063×10^{-3}
21133.9	3,44	12,45	$R(44)$	1.440×10^{-3}
21134.1	3,41	12,41	$Q(41)$	1.434×10^{-3}
21134.3	6,46	17,45	$P(46)$	6.537×10^{-4}

TABLE III. A-band laser lines observed with a 6590-Å pump.[a] λ pump (air) = 6593.86 Å; λ pump (vacuum) = 6595.69 Å/15161.4 cm^{-1}.

λ air	λ vac	ν vac (cm^{-1})
7849.30	7851.47	12736.5
7856.93	7859.1	12724.1
7897.40	7899.58	12658.9
7897.90	7900.08	12658.1
7917.83	7920.01	12626.2
7929.47	7931.66	12607.7
7936.97	7939.16	12595.8
7974.74	7976.94	12536.1
7976.57	7978.77	12533.3
7990.91	7993.11	12510.8
7996.60	7998.81	12501.9
8008.40	8010.61	12483.4
8036.50	8038.72	12439.8
8039.31	8041.53	12435.4
8044.47	8046.69	12427.5
8053.66	8055.88	12413.3
8056.11	8058.33	12409.5
8069.43	8071.66	12389.0
8080.50	8082.73	12372.1

[a] Wavelength accuracy to within ±0.1 Å.

Torr. With a total cell pressure of 35 Torr we estimate that 27 Torr of the He buffer gas was mixed in the sodium vapor column.

Success at optical pumping in the B-band pump region led to experiments to pump the A band of the molecule. Prior to these experiments, we determined if other doubled Nd:YAG lines would also pump Na_2 transitions. We found that 5320 Å showed very little absorption and that 5620 Å showed strong absorption and defocusing, but no molecular emission. The evidence pointed to an atomic interaction with the pressure-broadened Na resonance lines. The 6594-Å line showed strong molecular absorption and led to laser emission on the A band. However, in this case the laser emission required the use of an optical cavity. We formed a cavity using 2- and 6-m-radius mirrors with reflective coatings from 6800 Å to 1.1 μm. The output coupling varied between 0.3 and 3%, and was considerably smaller than optimum for these high-gain A-band transitions. The A-band optical-pumping experiment is illustrated in Fig. 2.

Na_2 A-band laser action was observed for pump wavelengths of 6594, 6691, and 6795 Å available from a Q-switched internally doubled Nd:YAG laser. Table III lists the observed laser wavelengths for the 6594-Å pump wavelength. An attempt was made to identify the A-band pump transitions and corresponding laser-emission vibrational-band lines for each of the above wavelengths using $X\,^1\Sigma_g^+$ molecular constants from Demtroder *et al.*,[5] and $A\,^1\Sigma_u^+$ molecular constants from Fredrickson, making the vibrational assignment corrections determined by Fredrickson and Stannard.[6] However, the fit was not accurate enough to make unambiguous line assignments. The more recent and accurate molecular constants obtained by Kusch and Hessel[7] at NBS should allow an identification of the pump bands and laser-emission lines.

The threshold pump energy for the A-band Na_2 laser was less than 1 μJ, as low as available from the Q-switched doubled YAG laser source. Laser action was observed over the sodium-vapor-column temperature range from 416 (0.014 Torr Na_2, 0.56 Torr Na) to 505 °C (0.175 Torr Na_2, 4.4 Torr Na) with 30—35 Torr of He buffer gas mixed in the sodium vapor. At intermediate sodium temperatures, typically 465 °C, sporadic laser action was observed at a buffer gas pressure of 162 Torr maximum with consistent laser action observed at the lower buffer gas pressures, 30—50 Torr. The output laser energy was only 2.35×10^{-3} μJ due to nonoptimum output coupling and possible losses at the cell windows due to sodium vapor fogging.

We have modeled the Na_2 molecular laser as a three-level system and have evaluated the cross sections, saturation intensities, and gain for A- and B-band transitions. We assume the pump-transition ground state and laser-transition final state are (v'',J'') levels in the X ground electronic state and the laser-transition initial state (v',J') lies either in the A or B excited Na_2 electronic states. For the isotropic molecular gas the stimulated-emission cross section is

$$\sigma_{SE}(\omega) = \frac{3}{4}\,\frac{\lambda_0^2 |\langle v'|v''\rangle|^2 g(\omega-\omega_0)}{\tau_{RAD}}, \tag{1}$$

where $g(\omega-\omega_0)$ is the normalized line-shape function, $|\langle v'|v''\rangle|^2$ is the Franck-Condon overlap factor for the transition $(v'J'\rightarrow v''J'')$, and τ_{RAD} is the radiative lifetime of the (v',J') emission band. The gain coefficient $g(\omega)=\sigma(\omega)\,\Delta N$ is related to the emission cross section through the population-inversion number density $\Delta N = [N' - N''(2J'+1)/(2J''+1)]$. The saturation intensity for cw optical pumping is defined by the expression

$$I_{SAT} = \frac{\hbar\omega_0}{2\sigma(\omega)T_1} = \frac{2}{3}\,\frac{\hbar\omega_0}{\lambda_0^2|\langle v'|v''\rangle|^2 g(\omega_0)}\,(1+\tau_{RAD}\gamma_{NONRAD}),$$

where $1/T_1 = 1/\tau_{RAD} + \gamma_{NONRAD}$ includes all the nonradiative inelastic relaxation processes out of the pump-transition upper state (v',J'). I_{SAT} above must be compared with the optical pump intensity or power density within the molecular linewidth.

For pulsed optical pumping, where we assume optical pumping pulses long compared to the radiative lifetime and a laser pump linewidth greater than the molecular linewidth, the peak saturation intensity per unit laser bandwidth is

$$\frac{I_{SAT}}{\Delta\omega_{LASER}} = \frac{hc}{\lambda_0^2|\langle v'|v''\rangle|^2}\,(1+\tau_{RAD}\gamma_{NONRAD}). \tag{3}$$

For flash-lamp pumping, the relevant saturation parameter is the saturation photon flux within the molecular linewidth per unit linewidth

$$\frac{I_{SAT}}{\Delta\lambda} = \frac{2\pi c}{\lambda_0^4|\langle v'|v''\rangle|^2}\,(1+\tau_{RAD}\gamma_{NONRAD}). \tag{4}$$

For a homogeneous Lorentzian line shape we multiply Eqs. (3) and (4) by $\frac{1}{2}\pi$ and for an inhomogeneous Gaussian line shape we multiply by $\frac{1}{2}(\pi/\ln 2)^{1/2}$.

We can now calculate the gain and saturation constants for optically pumping the A and B bands of the Na_2 molecule. For the B band with λ_p at 0.473 μm and λ_{laser} at 0.54 μm the Doppler widths are $\Delta\omega_D = 12.42$ GHz and $\Delta\omega_D = 10.88$ GHz at 600 °C. The B-band radiative lifetime is[8-10] $\tau_{RAD} = 6.5$ nsec. For strong laser transitions the Frank-Condon factor is typically $|\langle v'|v''\rangle|^2 \sim 0.1$ [11] so that the stimulated-emission and absorption cross sections are $\sigma_{EM}(\omega) = 2.90\times10^{-12}$ cm^2 and $\sigma_{ABS}(\omega) = 1.95\times10^{-12}$ cm^2. These cross sections are very large even compared to the 10^{-16}-cm^2 Rhodamine 6G dye laser cross section.

In a similar way, we can evaluate the Doppler width and gains for the A band. The Doppler widths at 0.8 μm (signal) and 0.67 μm (pump) are $\Delta\omega_D = 7.35$ GHz and $\Delta\omega_D = 8.77$ GHz. The corresponding cross sections are $\sigma_{EM}(\omega) = 5.11\times10^{-12}$ cm^2 and $\sigma_{ABS}(\omega) = 3.00\times10^{-12}$ cm^2, where $\tau_{RAD} = 12$ nsec.[12]

For the B-band saturation intensity in the Doppler-broadened operating region we find $I_{SAT} = 16.6$ W/cm^2 for pumping strong transitions at 0.473 μm. This value agrees well with a previous measurement taken at 0.488 μm using a cw argon-ion-laser source. Optically pumping the strong B-band transition ($v'' = 3 \rightarrow v' = 6$) $Q(43)$ at 0.488 μm we measured a saturation intensity of 28.5 W/cm^2 compared to a calculated 21.4 W/cm^2 based on a Franck-Condon factor of 0.068.[11] For B-band flash-lamp pumping Eq. (4) gives

$$h_{SAT}/\Delta\lambda = 2.68\times10^{21} \text{ photons/cm}^2\,\text{sec}\,\text{Å}^{-1},$$

with a Doppler linewidth of

$$\Delta\lambda_D = (\lambda_0^2/2\pi c)\Delta\omega_D = 0.015 \text{ Å}.$$

The saturation photon flux to pump the entire molecular line is

$$h_{SAT} = 3.94\times10^{19} \text{ photons/cm}^2\,\text{sec},$$

which is comparable to the photon flux available at 0.47 μm from high-pressure xenon flash lamps in tightly coupled elliptical cavities.[13] The A-band saturation-intensity values for optically pumping strong transitions in the Doppler-broadened regime are approximately one-fourth of the B-band values.

In the pressure-broadened regime with He as the buffer gas we expect the saturation intensity to increase. Assuming that single He-Na_2 collisions dominate the transverse "elastic" relaxation processes and the nonradiative longitudinal "inelastic" relaxation processes at moderately high He pressures, the effective pressure-broadened linewidth becomes

$$\Delta\omega_L = \frac{1}{T_1} + \frac{2}{T_2} = \gamma_{NONRAD} + 2\gamma_{ELASTIC},$$

assuming $\gamma_{ELASTIC} = \gamma_{NONRAD} = \sigma v_{He}$,[14] gives

$$\Delta\omega_L = 0.273 \text{ GHz/Torr}$$

and

$$\gamma_{NONRAD} = 0.09 \text{ GHz/Torr},$$

using $\sigma = 100$ Å^2 as the total inelastic cross section.[14] Hence Eq. (3) gives a pumping saturation intensity per bandwidth for the Lorentzian pressure-broadened line of 553.9 W/cm^2. At 760 Torr of He at $T = 600$ °C we find $I_{SAT} = 720$ kW/cm^2. This saturation intensity compares favorably with that of Rhodamine 6G and thus indicates that Na_2 may operate as a flash-lamp-pumped laser source with buffer gas broadening if other quenching mechanisms do not spoil the inversion.

Finally, we can calculate the Na_2 gain for saturation optical pumping with a laser source. For the strong B-band pump transitions listed in Table II, the average pump-transition ground-state population is 2.10×10^{13} cm^{-3} at $T = 600$ °C. Assuming an empty laser-transition final state, saturation pumping will achieve a maximum inversion density of 1.05×10^{13} cm^{-3} which gives an unsaturated gain of 30.5 cm^{-1} at $\lambda = 0.54$ μm. The measured superfluorescent output energy corresponds to a photon flux at least an order of magnitude below the calculated laser saturation intensity of 2.32×10^{19} photons/cm^{2}sec, so we may neglect gain saturation. Hence the experimentally determined gain of 4.07 cm^{-1} indicates an achieved population inversion approximately 13.5% of the maximum possible. Comparing the number of superfluorescent photons with the above population inversion indicates that nearly 100% of the inversion was dumped into the output pulse.

In conclusion we have shown that superfluorescence gain and laser action is readily obtained in an optically pumped Na_2 dimer on both the A and B emission bands. These results suggest that Na_2 is a promising candidate for other pumping schemes such as flash-lamp pumping or discharge pumping. Furthermore, Na_2 is representative of the other alkali molecular systems which have molecular emission bands extending across the visible to the near-infrared spectral region.

ACKNOWLEDGMENTS

The authors wish to acknowledge the support of the Office of Naval Research and the Air Force office of Scientific Research during the course of this work.

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1. M. Henesian and R. L. Byer, "Optically Pumped Na_2 Molecular Laser," presented at the CLEA conference, Washington, D. C., 1975.
2. M. Henesian, R. L. Herbst, and R. L. Byer, "Optically Pumped Superfluorescent Na_2 Molecular Laser," J. Appl. Phys. 47, 4 (April 1976).

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4. TITLE (and Subtitle) Optical Pumping of Molecular Gases		5. TYPE OF REPORT & PERIOD COVERED Interim 1 Apr 75 to 31 Mar 76 6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) Robert L. Byer		8. CONTRACT OR GRANT NUMBER(s) F44620-74-C-0039 NEW
9. PERFORMING ORGANIZATION NAME AND ADDRESS Edward L. Ginzton Laboratory W.W. Hansen Laboratories of Physics Stanford University, Stanford, CA		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 9751-02 61102F
11. CONTROLLING OFFICE NAME AND ADDRESS AFOSR/NP Bldg 410, Bolling AFB, DC 20332		12. REPORT DATE April 1976 13. NUMBER OF PAGES 37
14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)		15. SECURITY CLASS. (of this report) UNCLASSIFIED 15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

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19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

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During the past year we have completed optical pumping studies of Na_2 and have initiated optical pumping studies of CO and Hg_2. Optical pumping of Na_2 shows that it is potentially an interesting quasi tunable, high gain dimer laser in the .52 -.81 pm spectra range. Our results show that Na_2 could possibly be pumped by a flashlamp or an electrical discharge. Measurements on optically pumping Hg_2 show that 2mJ of .266 pm radiation is needed to achieve threshold on the .33 pm band. Experiments are in progress to achieve lasing in Hg_2.