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Cambridge, Massachusetts 02139

Application of Gas Lasers to Studies of Fundamental Molecular and Atomic Processes

ANNUAL TECHNICAL REPORT #3

for combined periods: January 1, 1970 to June 30, 1970
July 1, 1970 to December 31, 1970

Under Supervision of
Principal Investigator: Professor Ali Javan
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Molecular excitation and relaxation mechanisms are under investigation by a number of techniques. A fluorescence technique for observation of coherent inversion of optical transitions has been used to study relaxation in CO_2. Studies of excitation and relaxation processes in transverse and longitudinally excited HF lasers are in progress and results on a transverse room temperature CO laser are also reported. Studies of the pressure broadening and shift of several CO_2 transitions are in progress using a standing wave saturation resonance technique.

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This progress report summarizes the ongoing research of the Massachusetts Institute of Technology Laser and Quantum Electronics Group, under ONR Contract #N00014-67-A-0204-0014 performed during the period January 1, 1970 to December 31, 1970. Research performed includes:

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Three Appendices are also included:

A. Observation of Intense Superradiant Emission in the High Gain Infrared Transition of HF and DF Molecules.

B. High Pressure Transverse Discharge CO Laser.

C. Fluorescence Induced by Coherent Optical Pulses.

I. Study of Excitation and Relaxation Mechanisms in the HF and CO Lasers

Molecular Relaxation Studies in HF Gas

Work has begun on assembling the necessary experimental apparatus for investigating vibrational and rotational relaxation processes occurring in pure, gaseous HF and in mixtures of HF and other non-reactive gases. At present, a high vacuum, HF handling system has been designed and assembled. The system is constructed entirely of Kel-F and Teflon plastics. These materials are inert to chemical attack by HF, and, therefore, the formation of impurities via chemical reaction is prevented. The system includes two cold traps for use in further purifying HF by successive distillation. Purity of the condensed sample can be ascertained by electrical conductivity measurements. Finally, a capacitance manometer, for precision pressure measurements was designed and fabricated. The manometer design is similar to that of a standard capacitance manometer, except that only non-reactive teflon materials were used for the interior of the sensing head.

Near Infrared HF Laser

A systematic investigation of the high energy, high gain pulsed HF laser is in progress. Initial experiments with a transverse discharge (TEA) HF, DF laser are described in a paper published in Applied Physics Letters and included as Appendix A of this report. Briefly, the laser is capable of high power, high energy (> 50m. j) pulsed output in the attractive 3 and 4μ regions.

The laser device is simple and compact, and utilizes relatively nontoxic gases. However, its spatially non-uniform excitation scheme and its high gain make single transverse mode operation difficult. To overcome these difficulties a short narrow bore, high pressure, longitudinal discharge laser has been designed and operated. It is capable of pulse energies of several milli joules and peak powers > 2kW. Preliminary experiments indicate its single mode behavior is superior to that of the transverse discharge laser. A comparison of the spectral characteristics of the two lasers is now in progress.

Long Wavelength HF Laser

A wide bore, longitudinal discharge HF laser, to be used in studying far infrared emission in HF, has been built and operated. Laser excitation is accomplished by electrically pulsing a flowing mixture of H_2 and a fluorinated gas species such as SF_6, Freon 14, or Freon 13. The device design is such that a wide range of laser operating conditions and gas mixtures can be investigated. In studying the laser output spectrum, a fore-prism-grating spectrograph combination has been found to be a convenient method of isolating near and far infrared laser lines.

Using this apparatus, we have observed intense, pure rotational lasing in all three of the above named fluorinated gases. Laser wavelength ranged from 9 to 15 μ; corresponding to rotational transitions from J=17 to J=30. Many of these laser wavelengths fall within the well-known 8 to 14 μ atmospheric window. Studies of the variation in laser spectral properties with gas pressure and gas mixtures have begun. Initial results have already

provided clues to the process leading to rotational population inversion.

CO Transverse Discharge Laser

Details of experiments with a transverse discharge CO laser have been assembled in a paper to be publied in the June IEEE Journal of Quantum Electronics. This paper is included as Appendix B. Further work on C.W. and pulsed room temperature CO lasers are scheduled to commence early this summer. These experiments will include a continuing investigation of molecular processes occurring in the CO Laser.

II. Influence of Molecular Relaxation in Coherent Optical Processes

The study of fluorescence induced by short pulses of radiation in CO_2 has been concluded and the results submitted for publication (see Appendix C). It has been demonstrated that one can observe the coherent excitation of an optical level in this way, even in a very weakly absorbing transition, and extract values of the matrix element and relaxation times.

Present efforts in the area of short pulse propagation and coherent interaction have shifted to an investigation of adiabatic rapid passage.[1] This technique has long been used in magnetic resonance to invert a spin system and study relaxation processes. Previous works in our laboratory on Stark spectroscopy of NH_3 and NH_2D has uncovered a number of transitions which can be swept through resonance with a CO_2 or N_2O laser transition. Because of their large absorption coefficient and tunability, these transitions make excellent candidates for the study of adiabatic rapid passage. These experiments are of interest both from the point of view of studying the propagation of chirped laser pulses through an absorbing or amplifying medium, and for the study of relaxation processes.

The laser and Stark cell are functioning properly and adequate absorbtion signals have been observed to make an attempt at observation of adiabatic rapid passage feasible in the near future.

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1. E. B. Treacy, Phys. Letters 27A, 421 (1968)

III. Molecular Studies Using Standing Wave Saturation Resonances

The study of the standing-wave saturation resonance in low pressure CO_2 is well underway. The effect, first observed by Freed and Javan,[1] is being studied in detail to determine the influence of collisions and intense laser fields on the detailed line shape. In the experiments being performed, the low pressure CO_2 absorption cell is placed outside the laser cavity and the CO_2 laser, which is spaced with 4 invar rods for rigidity and thermal stability, is mounted firmly on a half-ton steel rail to minimize vibration. To further minimize laser jitter we have found it necessary to run the laser under very slow flow conditions. This has been done without the loss of laser intensity as would have been the case with a sealed off laser. Data is being taken on several of the CO_2 laser lines to determine the pressure shifts and saturation parameters of each transition. The signal to noise obtainable by this technique for locking a laser to this narrow resonance is demonstrated in Figure 1.

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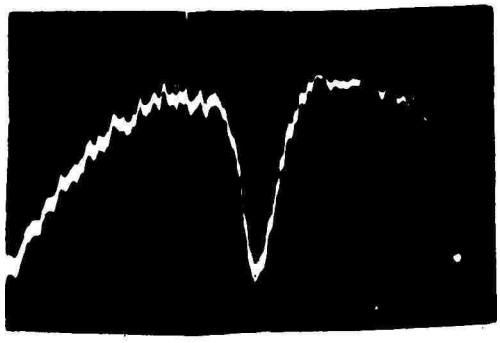
1. C. Freed and A. Javan, App. Phys. Letters 17, 53 (1970)



Standing wave saturation resonance in CO_2 gas. Full width of resonance is ~ 1 MHz.

APPENDIX A. OBSERVATION OF INTENSE SUPERRADIANT EMISSION IN THE HIGH GAIN INFRARED TRANSITIONS OF HF AND DF MOLECULES

J. Goldhar, R. M. Osgood, Jr. and A. Javan.

ABSTRACT

Intense stimulated emission (superradiance) is obtained in several high gain transitions of HF and DF molecules. The narrowing of the linewidths of the high gain transitions are observed in detail. Pulsed energy and peak powers are comparable with those obtainable in a similar transverse CO_2 laser.

This letter reports the observations of intense stimulated emission (superradiance) in a number of high gain, rotational-vibrational transitions of HF and DF molecules in the 2.7μ and 3.8μ range of wave-length respectively.[1,2] The vibrationally excited molecules are produced with a transverse pulsed discharge in a flowing mixture of molecular hydrogen (or deuterium) and SF_6 gas, at relatively high pressure. The spectral distribution of the individual high gain transitions have been studied with a pneumatically tuned Fabry-perot interferometer. Each of the superradiant transitions is found to show considerable line-width narrowing due to substantial amplification of the traveling optical wave. In addition, it is found that in the presence of a regenerative optical feedback (i.e . with the device used as a laser with an optical resonator), the laser oscillation on each line occurs mainly in a single resonator mode. With regard to the obtainable peak power and the pulse energy, the performance of the system is competitive with that obtained at 10.6μ using a CO_2 laser excited by means of a similar transverse pulsed discharge.

In the experiment, the laser configuration consisted of a 50 cm long transverse discharge [3] with 101 equally spaced pin electrodes arranged in a row facing a long cylindrical brass, anode across a 2.5 cm gap. A 0.01 μF capacitor at high voltage is discharged across the tube through a 1000 ohm resistor in series with each pin. The discharge tube was terminated with CaF_2 Brewster windows. Gases were introduced into the system by means of standard needle valves and flow was maintained with a 10 CFM fore pump.

In the experiment, most of the detailed observations were made with the hydrogen gas. Typically, the partial pressure ratio of SF_6 to H_2 was 10:1. In this system, the vibrationally excited HF molecules are formed through chemical reaction [4] of the fluorine atoms, (which exist as a byproduct of the SF_6 dissociation), and the molecular hydrogen. As noted earlier, [2] this reaction is 31.7 kcal/mole exothermic for HF and DF, and hence capable of producing vibrationally excited HF up to the vibrational level v=3/(and up to v=4 in DF).

The distribution of infrared output radiation among the various rotation-vibration transitions in HF, is found to be dependent on the operating conditions such as flow rate, partial and total pressure, and applied voltage. If the device is operated as a laser with a regenerative optical feedback, at relatively low pressures of SF_6 and H_2 and low applied voltage, (below about 10k.v.), the output is distributed among several rotation-vibration transitions of each of the three v=3→2, 2→1 and 1→0 vibrational bands.[5] At the elevated pressures and for the applied voltages in the regions of about 15 kilovolts (or higher), the laser output spectrum tends to peak in the v=1→0 transition with P (4) the most energetic line. This behaviour indicates that at the elevated pressures and voltages, (where the HF population is appreciable), the HF molecules in the v→1 states decay rapidly to the v=1 state, causing a build-up of population in the v=1 state, which apparently decays at a slower rate.[6]

An important characteristic of this system is its capability of producing very high gains leading to intense stimulated

emission without regenerative optical feedback (superradiance). In analyzing the output spectrum of the superradiant device, it was most essential to guard against unwanted optical feedback introduced by small reflections or scattering from auxiliary optical components present in the path of the output beam. This was done by introducing heavily absorbing filters placed at an angle in the path of the output beam and appropriately misaligning the various components to prevent the feedback. Such precautions were particularly important when a folding mirror was placed at one end of the amplifier to obtain double pass amplification at the output. Fig. 1 shows the method used to monitor the presence of an unwanted optical feedback. A sample output was provided by placing a beam splitter on the amplifier axis near the folding mirror. When the light emanating from the output end of the amplifier was interrupted, the presence of any feedback exterior to the amplifier could then be detected as a signal appearing at the sampling output.

In the high pressure region (about 200 torr SF_6 and about 20 torr H_2), it was possible to obtain intense stimulated emission on the P(4), v=1→0 and P(3), v=2→1 transitions with the amplifier used in a single pass; the P(4), v=1→0 was generally found to be more intense.

A pneumatically scanned Fabry-Perot interferometer with a 8.26 cm mirror separation was used to analyze the spectral distribution of each high gain transition. In this system, the output of the interferometer was detected through a small pinhole with an infrared detector. The interferometric analysis of the spectral distribution in each superradiant transition showed a

full-width at half maximum below 110 MHz. The 110 MHz limit was instrumental, determined by the finesse of the Fabry-Perot. This result was obtained with the amplifier used in both the single and double pass configurations.

An additional noteworthy behaviour of the system is that under the superradiant conditions, it is possible to obtain stimulated emission with sufficient intensity to cause amplifier saturation even for a single pass amplifier. This was observed by noting that at the limit of highest gain, the double-pass amplifier output on the P(4), $v=1\rightarrow0$ transition was only about 20 times larger than that obtained in the single pass. Furthermore, using the feedback monitor described above, the sampled signal of the double pass amplifier with no feedback was compared with that of the amplifier used as a laser with a complete regenerative feedback. The power level in the latter case was found to be only a few times higher than that of the double pass amplifier

Under the high pressure condition where the amplifier gain is near optimum, the spontaneous emission linewidth is essentially due to collision broadening. The exact magnitude of this broadening is not as yet known for the HF transitions. However, in the presence of about 200 torr SF_6 pressure, the spontaneous emission line width can be assumed to be at least about 3000 MHz. Accordingly, the observed 110 MHz line width limit corresponds to a line narrowing of 30 or greater. In the absence of saturation, this factor is known to be given by the square root of GL where G is magnitude of the gain per unit length and L is the amplification path length. However, it is important to guard against hasty application of this relationship in obtaining an accurate measure

of the amplifier gain in this system. For instance, it is probable that the presence of each pin electrode in the amplifying medium can cause appreciable scattering of the infrared emission to introduce sufficient feedback to cause additional narrowing of the line profile.[7] Furthermore, the saturation effect is also expected to play an appreciable effect on the line-shape.

The frequency spectrum of each oscillating line was also analyzed when the device was used as a laser with two aligned mirrors providing complete regenerative feedback. Interestingly, the frequency spectrum of each oscillating line, as averaged over a number of pulses, was found to be distributed within a frequency interval below about 70 MHz resolution limit of the Fabry-Perot[8]. In this case, a much broader spectrum is expected because of the possibility of the laser oscillating on several longitudinal modes of the laser resonator which was 118 cm long (corresponding to 130 MHz mode frequency spacing). The narrow line-width for each oscillating line was observed while the gas pressure and the applied voltage and hence the gain in the various transitions were varied over a wide range. (The lowest total gas pressure was somewhat less than 10 torr). While it is tempting to attribute this persistent behaviour to the high gain property of the medium, it is more likely that the effect originates from mode coupling in the presence of collision broadening. For a collision broadened line, the saturation effect decreases the gain uniformly over the whole line profile leading to mode competition . In general, however, complete single moding is not always expected for a collision broadened line due to the so called " spatial hole burning" effect. This effect occurs

in a region of space within the resonator where the crest of the standing wave for one mode falls near the node of another. This would allow coupling of both modes to the amplifying medium when the molecular mean free path is appreciably below one wavelength - a condition valid for a fully collision broadened line.

In our system, however, the spatial hole burning of this type may be smeared because of rapid density fluctuations and the acoustical waves caused by the high current discharge pulse[9]. This would inhibit multimoding on, say, several longitudinal modes but would allow simultaneous oscillation on a longitudinal and a transverse mode having appreciably different intensity distribution in the direction transverse to the laser axis. The latter possibility was verified by using an appropriate pair of curved laser mirrors to introduce transverse modes separated in frequency by tens of MHz from the corresponding longitudinal modes. With a fast response infrared detector, it was possible to obtain beat notes in the appropriate frequency regions dependent, as expected, on the degree of aperturing of the laser resonator. This result indicates the possible simultaneous presence of one longitudinal and several transverse modes at frequencies falling within the 70 MHz limit of the Fabry-Perot.

Study of the output power obtainable from this sytem has revealed an important practical property; namely, that the system is potentially capable of supplying considerable energies and high peak powers in each pulse. The optimum power and efficiency was found to be an increasing function of the applied voltage. At the 20 kv limit of the available high voltage supply, it was possible to obtain 16 kw peak power (corresponding to 0.6% efficiency) in a pulse of 700 ns duration. This was obtained in

a mixture of SF_6, H_2 with partial pressures of 300 and 10 torr respectively. This result suggests that by increasing the applied voltage and scaling the volume and the gas pressure, it is possible to obtain much larger output powers, competitive with those reportedly obtainable in the transverse discharge CO_2 laser.

With the D_2 gas used instead of H_2, it was possible to obtain intense stimulated emissions on the DF transitions in a manner similar to that observed in HF, as described above. With the amplifier used as a laser with regenerative feedback, the DF oscillations appeared over several transitions in the v=4→3; v=3→2, v=2→1, and v=1→0 bands. In the high pressure region, the lines capable of intense superradiant emission were P(5) and P(6) of both the v=2→1 and V=3→v=2 bands. More detailed studies of the pressure dependence of the DF spectrum are now in progress.

In conclusion, we would like to point out that the high pressure transverse gas discharge provides a general method of extracting high peak powers and pulse energy for many other gas laser transitions. For instance, we have recently observed intense laser oscillations in the 5μ bands of Pure CO excited by a transverse gas discharge at relatively high pressures. Detailed study of the system will be published separately [10].

We would like to acknowledge Dr. F. Zernike for making available the Fabry-Perot flats used in the experiment. We would also wish to thank Mr. Ronald McNair and W. L. Ryan for their expert technical assistance. Lastly, we are grateful to Prof. E. V. George and Dr. R. Carbone for numerous helpful discussions.

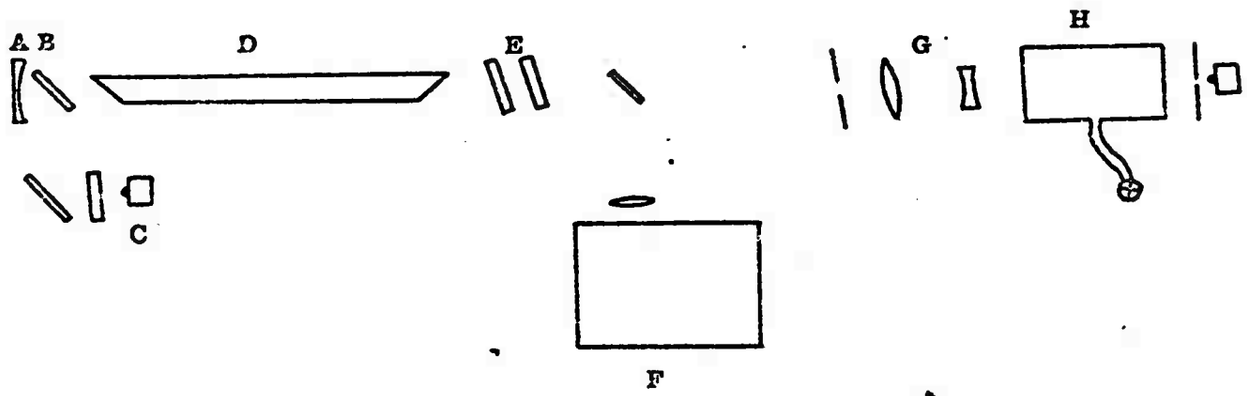
1. Laser oscillations in the 2.7μ and 3.8μ bands of HF and DF molecules formed chemically in a gaseous discharge, were separated sometime ago by T. F. Deutsch, Appl. Phys. Letters, 10, 234, 1967. Similar results obtained in a transverse discharge at the 2.7μ band of HF have recently been reported by M. C. Lin and W. H. Green, J. Chem. Phys. 53, 3383, 1970. See also C. J. Ultee, IEEE J. of Quantum Electronics, QE-6, 647, 1970. This publication shows the possibility of obtaining large gains in an electrically pulsed, chemical HF laser.
2. For chemical HF, DF lasers produced in a supersonic flowing mixture of D_2 or H_2 with thermally heated SF_6, see D. J. Spencer, H. Mirels, and T. A. Jacobs, Appl. Phys. Letters 16, 284, 1970. For the application of flash photolysis, see J. H. Parkes and G. C. Pimentel, J. of Chem Phys., 51, 91, 1969.
3. A. J. Beaulieu, Appl. Phys. Letters, 16, 504, 1970.
4. Parkes and Pimentel, J. of Chem. Phys., 51, 91, 1969.
5. The transitions in the $v=3\rightarrow2$ and $v = 2\rightarrow1$ bands ranged from P(3) to P(6) with P(3) generally being more intense. In the $v=1\rightarrow0$ band the transitions P(4) through P(6) were observed with P(4) being the most intense.
6. In this process, the rapid decay of the HF molecules in the $v>1$ states to the v=1 will be dominated by v-v type collisions with HF molecules in the v=0 state. However, the slower decay of the v=1 state will result from collisions with H_2 and SF_6 molecules.

7. Under the high gain conditions, it was possible to trigger the amplifier into oscillation with a diffuse regenerative feedback. For instance, by holding a sheet of paper at the output end of the folded amplifier, it was possible to obtain strong oscillations.
This could be detected by means of the feedback monitor (see Fig. 1). For a discussion of non-resonant feedback and its implications see R. V. Ambartsumyan, N. G. Basov, P. G. Kryukov and V. C. Letokhov JETP 24, 481 1967; and R. V. Ambarsumyan, P. G. Kryukov and V. C. Letokhov JETP 24, 1129, 1967.

8. When the device was used as a laser with regenerative feedback, the interferometer could be better aligned with respect to the incident beam without introducing feedback into the laser. Accordingly, a better system finesse was then obtained.

9. An additional source of the smearing of the spatial hole burning may also arise from a rapid variation of the refractive index during the build up of radiation within the resonator causing a wavelength sweeping of each mode. Such an index change may arise from molecular dissociation due to the current pulse and the build-up of population in the high gain amplifying transitions contributing to the refractive index.

10. Osgood, Goldhar and McNair to be published in Journal of Quantum Electronics.


A B
D
E
G
H
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CAPTIONS

Fig. 1: Experimental apparatus for Analyzing the Behaviour of the Superradiant Device.

A) Folding Mirror; B) Sampling;

[see text for explanation]

C) Sampling detector with filter beamsplitter;

D) Discharge tube; E) Absorbing Filters

F) Monochrometers G) Collimator

H) Febry-Perot Interferomater with pressure scanning valve, pinhole and detector.

APPENDIX B. HIGH PRESSURE, TRANSVERSE DISCHARGE CO LASER

R. M. Osgood, Jr., J. Goldhar and R. McNair

ABSTRACT

Experiments with a high pressure, transverse discharge CO laser are described. The pulsed laser output was observed to occur on several CO rotational-vibrational transitions in bands from v=6 → v=5 to v=11 → v=10. A study of the laser delay time versus CO pressure is presented and its relation to two inversion schemes are discussed in detail.

In an earlier publication we reported, briefly, oscillation on several vibrational-rotational transitions in a pulsed transverse discharge CO laser. We give here a more detailed description of this experiment.

The discharge tube used was essentially the same as that described in Ref. 1 for the superradiant HF laser. Because of the low gain of the CO laser, it was necessary to use a high Q cavity consisting of two gold mirrors, one of which had a 1mm dia output coupling aperture. The laser was normally operated with pure CO at pressures from 2 torr to 1/3 of an atmosphere. Addition of He (or N_2) to the discharge increased the laser peak power only at pressures $\lesssim$ 20 torr. With a CO pressure of 150 torr, a peak power of 100 watts and a pulse width of 2μ-sec. was observed.

A study of the laser spectroscopy revealed that each pulse was in reality a superposition 6 individual pulse trains; one for each of the vibrational bands observed in oscillation. Table 1 gives a brief summary of the transitions observed under optimum power conditions. The delay time, before oscillation occurred for each pulse train, was observed to increase in the order $v = 6 \rightarrow v = 5$ to $v = 11 \rightarrow v = 10$. This sequential time ordering was nearly identical to that observed by Patel [2] in a low-pressure, longitudinal discharge, pulsed CO laser several years ago. The delay time for each transition was found to vary with pressure and capacitor voltage. Fig. 1 shows a typical plot of variation of delay

time with tube pressure for the P(11) $v = 6 \rightarrow v = 5$ transition. Note that the decrease in slope of the curve as the delay approaches 1μ sec. is to be expected on the basis of a finite pulse rise times for the minimum gains expected in this laser. Finally it was found that an increase in the total pulse width occurred with decreasing pressure. Thus, the delay time for oscillation to occur on each vibrational band also varied with pressure.

Due to the fact that our high-pressure CO laser contains only a single gas, it offers an excellent opportunity to understand the excitation mechanism of a transverse discharge molecular laser at a fundamental level. For the E/p ratios seen in our CO discharge, Nighan [3] has recently shown that virtually all of the electrical power is dissipated in populating the CO, lower vibrational levels* and $a^3\Pi$ metastable state. As shown below, both of these two processes may provide the basis for an inversion.

Considering first the $a^3\Pi$ state, we note that several recent measurements of its radiative lifetime have yielded values of the order of a few milliseconds [4]. This is a factor of 100 larger than that required by the laser delay time measurement, and thus radiative decay from the $a^3\Pi$ state can be ruled out as an inversion mechanism. However, the collisional decay rate via electronic to vibrational energy transfer is not known, and the value of 20μ-sec.-torr as seen in the delay time measurements seems reasonable.† This

quenching process would be expected to have the inverse pressure dependence shown in Fig. 1.

In studying direct electron impact excitation of the CO vibrational levels, Schulz [6] found that the cross section for excitation from the V = 0 to the V $\leq$ 8 was large ($\sim 10^{-15}$ cm^2). The fact that vibrational excitation occurred during the current pulse was verified by observing the spontaneous 5μ fluorescence with the onset of the current pulse. Population of the vibrational upper levels, to the degree that laser oscillation occurs, can thus take place only after many vibrational-vibrational exchanges. Sharma has recently calculated the time for exchange of a vibrational quantum in CO assuming dipole-dipole interaction and found it to be 10μ-secs.-torr [7]. The delay times seen in our experiment are consistent with such a time scale. In addition, one would expect an inverse pressure dependence, as found in the laser delay time experiment. A variation of delay time with voltage is expected since only collisions between excited species are effective in pumping.

By noting which transitions within the v = 7 → v = 6 band are seen in laser oscillation, and, assuming a thermal rotational distribution, one can determine the effective vibrational temperature between the v = 7 and v = 6 levels to be greater than 10,000°K [2]. Similar or even higher vibrational temperatures are seen in laser transitions from other vibrational bands. A major problem with the above pumping mechanism then appears to be in accounting for the

lack of laser oscillation in the lower vibrational bands (particularly immediately after the current pulse). This problem may be resolved by considering the exothermicity of the vibrational exchange process [8],

$$CO\ (v=n) + CO\ (v=m) \rightarrow CO(v=m-1) + CO\ (v=m+1) + \Delta KE$$

where $n < m$.

As has been shown analytically by Brau, et al [9] and experimentally by Taylor [10], this process may lead to a quasi-equilibrium such that the effective vibrational temperature in the low vibrational states will be as low as 3000°K, while in the upper levels it may exceed 15,000°K. The fact that such a collisional process has been used to explain the anomalous vibration level distributions in non-electrically excited CO lasers [11], [12] is a further argument for the existence of nonequilibrium vibrational pumping in our laser. Finally, both the ordering and the magnitudes of the delay times for each vibrational bands suggests strongly that the laser levels are being pumped by the lower vibrational levels. Unfortunately, however, the interaction of the levels through laser-induced cascading prevents a clearcut interpretation.

In conclusion, we have shown that it is possible to obtain strong laser oscillation in a room temperature, high pressure CO discharge of transverse configuration, and that the delay times in this laser follow a trend similar to those found

earlier in a low-pressure, longitudinal discharge CO laser. Also, while it is tempting to ascribe the laser inversion to nonequilibrium vibrational pumping, uncertainty about the magnitude of the electronic to vibrational relaxation of the CO $a^3\Pi$ state prevents a firm conclusion from being drawn. We note that a careful study of the CO infrared spontaneous emission with time resolved spectroscopy should provide a definite answer.

We would like to acknowledge the invaluable guidance and support of Prof. A. Javan. In addition, we wish to thank Drs. R. Center, R. Sharma and V. George for many fruitful discussions and Drs. W. Borst and E. Zipf for supplying a preprint of their paper prior to publication.

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Band	v=6→ v=5	v=7→ v=6	v=8→ v=7	v=9→ v=8	v=10→ v=9	v=11→ v=10
Transitions	P(9) P(11) P(12) P(14)	P(10) P(11) P(12) P(13) P(14)	P(9) P(10) P(12) P(14)	P(9) P(10) P(11) P(12) P(13)	P(8) P(9) P(10) P(11) P(12) P(13)	P(8) P(11) P(13)

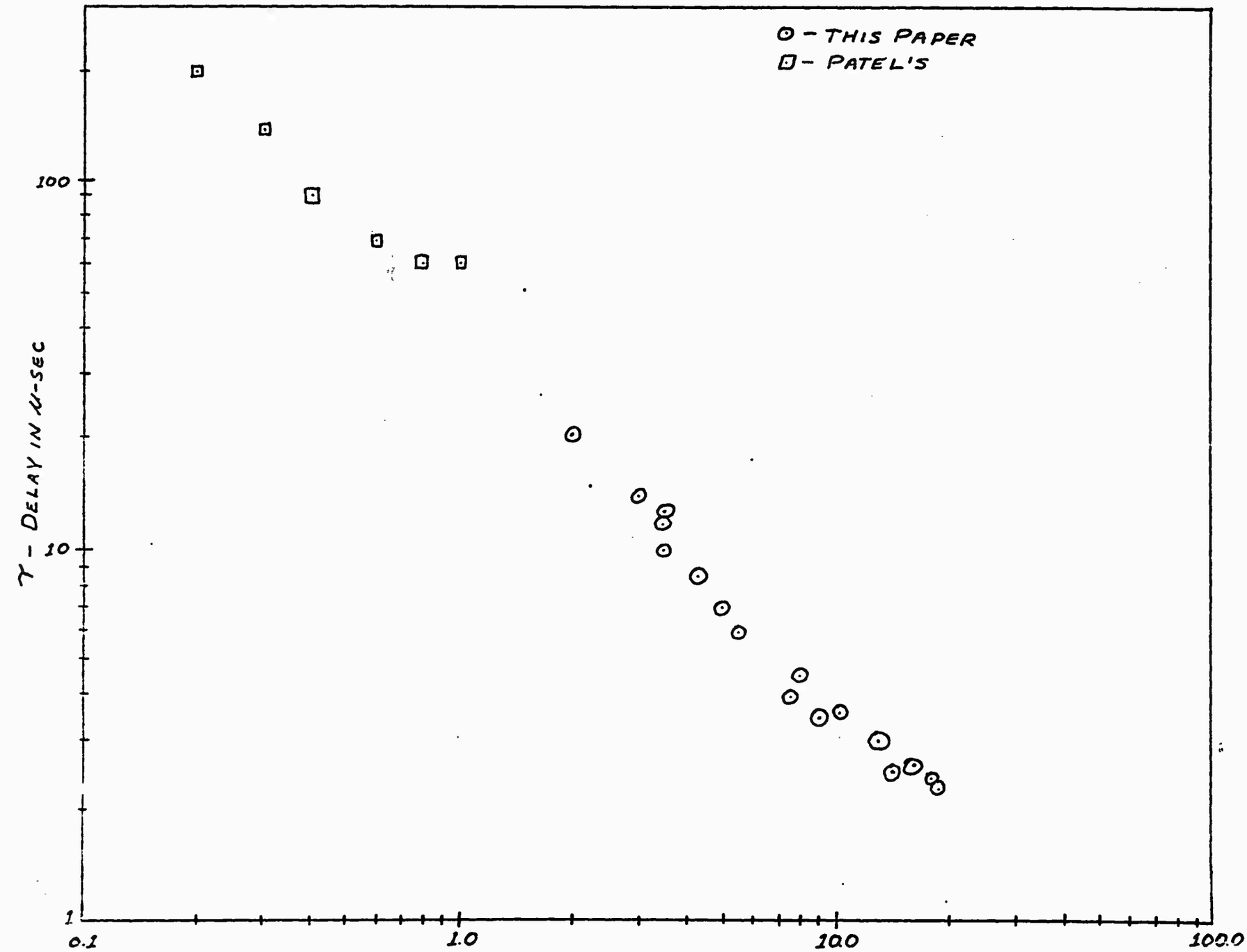
List of Captions

Fig. 1: Plot of Delay Time, "τ" (for lasing on the transition $v=6 \rightarrow v=5$ P(11)) versus CO Pressure, "P". Discharge Voltage is 15kv. For comparison delay time measurements for the same transition from Fig. 7 of Ref. 2 are included. The slope of these data give $\tau \propto \frac{1}{P}$.

Table 1: Observed transitions in the transverse discharge CO laser for typical low pressure operating conditions; 15kv discharge voltage, 8 torr CO pressure.

*Nighan's data concerns only c.w. discharges. However, using the appropriate electron collision cross sections, one finds that the mean free time between collisions (for an electron) in our laser is much less than the duration of the current pulse. Hence, our laser pulsed discharge will be similar to Nighan's c.w. model.

†While vibrational-electronic energy transfer has not been examined for CO-CO collisions, it has been studied for a Hg-CO system [5]. Collisional energy transfer from mercury metastables was found to be responsible for excitation of CO to high-lying vibrational levels ($v \sim 7$). Note, however, the large cross section for transfer was due to formation of a temporary chemical complex between Hg and CO.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τ - DELAY IN μ-SEC
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PASSIVE FACTOR

APPENDIX C. FLUORESCENCE INDUCED BY COHERENT OPTICAL PULSES

H. P. Grieneisen, N. A. Kurnit and A. Szöke

ABSTRACT

A coherent electromagnetic field, resonant with an optical transition, drives the molecules of a gas into the upper and lower states alternately, unless relaxation is too fast. The observation of this coherent excitation of optical levels by fluorescence measurement is discussed and experimental results are presented. The 4.3μ fluorescence of CO_2 gas excited by a 10.6μ laser is studied; values for the dipole moment and relaxation times are obtained in good agreement with those obtained by other methods.

Studies of photon echoes,[1] adiabatic rapid passage,[2] transient nutation,[3] and self-induced transparency[4] have extended into the optical region many of the methods used in magnetic resonance for probing relaxation processes. This letter presents results on a related, sensitive method of examining relaxation effects by the measurement of fluorescence[5,6] excited by a coherent, monochromatic optical pulse in an optically thin sample.[7] In this case, a simple and direct correspondence between the analysis and the experimental results emerges; complications from nonlinear propagation effects[3,4] do not appear.

We consider an atom or molecule in a gas with three energy levels of primary interest. The system is initially in state 1; a resonant optical pulse excites the system coherently to state 2, and the system subsequently decays to state 3 (possibly identical to state 1) while radiating spontaneously. The optical electric field of the running wave pulse, at the position of the atom, x, is described by:

$$\vec{E}(x,t) = \vec{\varepsilon}\mathcal{E}(x,t)\cos(\omega t-kx) \tag{1}$$

where $\vec{\varepsilon}$ is the polarization vector, $\mathcal{E}(x,t)$ is a slowly varying envelope, and $\omega\approx\omega_{12}$ is the optical carrier frequency, possibly also varying slowly in time. During the pulse the equation of motion of the density matrix ρ can thus be approximated by that of a two-level system:

$$\dot{\rho} = -\frac{i}{\hbar}[H,\rho] - \begin{pmatrix} (\rho_{11}-\rho_{11}^{eq})/T_1 & \rho_{12}/T_2 \\ \rho_{21}/T_2 & (\rho_{22}-\rho_{22}^{eq})/T_1 \end{pmatrix} \tag{2}$$

where the Hamiltonian $H = H_o - \vec{\mu}_{op}\cdot\vec{E}(x,t)$ consists of the unper-

turbed part, H_o, having the energy levels 1, 2, and the interaction of the optical field with the electric dipole moment of the atom, $\vec{\mu}_{op}$. The relaxation is treated phenomenologically (Bloch equations), it is characterized by the decay time of the energy, T_1, and that of the coherent dipole moment, T_2. The relaxation can be caused by radiative processes, collisions,[8] or diffusion. A more detailed account is beyond the scope of this letter. Eq. (2) can be integrated (as a rule numerically), with the initial condition $\rho_{ij}(t=0) = \rho_{11}^{o}\delta_{i1}\delta_{j1}$, to give the value of $\rho_{ij}(t=t_a)$ after passage of the pulse. If for simplicity we assume that the lifetime of the excited population is much longer than T_2, the decay of $\rho_{22}(t>t_a+T_2)$ can be considered separately since $\rho_{12}\to 0$. Thus the fluorescence intensity probes the upper state population at the end of the pulse. We can illustrate the expected results in some simple cases:

1) No relaxation, field on resonance for all the atoms. Eq. (2) can be integrated immediately in the rotating wave approximation to yield:

$$\rho_{22}(t_a) = \rho_{11}^{o}\sin^2\int_{o}^{t_a}(\mu\mathcal{E}(x,t)/2\hbar)dt \qquad (3)$$

where $\mu = |(\vec{\varepsilon}\cdot\vec{\mu}_{op})_{12}|$. This is a dramatic effect. Increasing the field (or lengthening the pulse) first increases, then decreases the observed intensity. The period of the modulation yields the value of the dipole moment μ. Also it is a direct measure of the quantity $\theta = (\mu/\hbar)\int_{-\infty}^{+\infty}\mathcal{E}(x,t)dt$, introduced by McCall and Hahn,[4] which has importance in propagation effects. As an optically thin sample changes the value of θ only slightly, this experiment can be used as a diagnostic tool in self-induced transparency and

related studies. The angle θ is also related to the pulse energy/unit area, S, by $\theta^2 = 8\pi\mu^2 S\tau_p/c\hbar^2$, provided we define the pulse width as $\tau_p = [\int_{-\infty}^{\infty} \mathcal{E}(x,t)dt]^2/\int_{-\infty}^{\infty} \mathcal{E}^2(x,t)dt$.

2) For fast relaxation, $T_1, T_2 \ll \tau_p$, the steady state solution applies. For a square resonant optical pulse one has

$$\rho_{22}(t_a) = \frac{\rho_{11}^o}{2} \frac{(\mu\mathcal{E}/\hbar)^2 T_1 T_2}{1+(\mu\mathcal{E}/\hbar)^2 T_1 T_2} \tag{4}$$

It can be seen that the fluorescence increases monotonically with pulse energy, first linearly, then more slowly. The saturation field measures the product T_1T_2.

In our experiment we deal with a spatially degenerate rotational-vibrational transition of CO_2. The measured fluorescence intensity is then proportional to

$$\bar{\rho}_{22}(t_a) = \int d(\Delta\omega) g(\Delta\omega) \sum_{M=-J}^{M=+J} \rho_{2M,2M}(\Delta\omega, t_a) \tag{5}$$

where $g(\Delta\omega)$ is a spectral distribution function (a Gaussian of width $\Delta\omega_D$ for Doppler-broadening).[9] In the case of spatial degeneracy, the selection rule $\Delta M=0$ for a linearly polarized field allows the transition to be treated as a collection of two-level systems, each one with its own resonance frequency and matrix element $\mu_M(J) = \mu_o(J)(1-M^2/J^2)^{1/2}$ for a P-branch transition.[10] Eq. (2) and the integral (5) have been evaluated for width $\Delta\omega_D/2\pi = 60$MHz and for various pulse shapes on a digital computer and results are presented in Fig. 1 for some cases of interest. The fluorescence intensity is plotted against laser field strength for various T_1, T_2 values. The salient features are: At long relaxation times (low pressures) there is modulation even for

high J transitions,[11] the modulation disappears at $T_2 \approx \tau_p$. The fluorescence intensity is always linearly proportional to the laser intensity (quadratic in $\mathcal{E}$) at low laser powers. For $T_1, T_2 \ll \tau_p$ it becomes proportional to the square root of laser intensity (linear in $\mathcal{E}$) when $(\mu\mathcal{E}/\hbar)(T_1T_2)^{1/2} \gtrsim 1$ and finally saturates when $(\mu\mathcal{E}/\hbar)(T_1/T_2)^{1/2} \approx \Delta\omega_D$.[12] The parameters μ, T_2, $(T_1T_2)^{1/2}$ can be estimated from the first minimum, the disappearance of the modulation, and the linear to square root transition region respectively.

Our experimental arrangement is shown in Fig. 2a. A 4m flowing CO_2-N_2-He laser (A), operating on the P(20) 10°0-00°1 10.6μ transition, with line selection provided by a diffraction grating (B), is Q-switched by a rotating mirror (C). Apertures (D) placed at both ends of the cavity each introduce ∿30% diffraction loss in order to eliminate off-axis modes and improve line selection, resulting in a pulse with good amplitude and shape stability (Fig. 2b). Frequency drift of the laser was corrected by moving the 90% reflecting laser output mirror (E) with a piezoelectric transducer (F). The laser gain was kept sufficiently low that detuning of the cavity mode by more than ∿4MHz from line center was readily evidenced by amplitude and shape changes in the laser pulse. The output of the laser passes through an amplifier tube (G) which provides a pulse of typically 2kW peak power and 250 n sec width at half maximum, a dimethyl ether absorption cell (H) utilized to vary the pulse intensity, and two CO_2 sample cells (I,J). It is then attenuated by filters (K) and focused by a lens (L) onto a Ge:Au detector (M) used to monitor the laser intensity and pulse shape. The 4.3μ

fluorescence from the 00°1 to 00°0 CO_2 level (see Fig. 2c) was observed with InSb detectors (N). The second sample cell (J), containing CO_2 at relatively high pressure (4 torr), was used to insure that nonlinearities observed in the low pressure sample were not due to undetected change in the laser frequency or pulse shape.

In order to obtain uniform intensity in the low pressure sample cell (I), a 5mm dia. aperture was placed inside the cell in the center of the beam. This, together with a 7mm aperture placed 4 cm behind it, limits the field of view of the detector to this central 4 cm region, a distance in which diffraction of the laser beam does not produce serious nonuniformities. These apertures also served to reduce a linear background signal believed to orginate from CO_2 adsorbed on the cell windows. The laser profile at the position of the first aperture was measured to be uniform to within 30%, which gives a sufficiently small field variation to avoid severe smoothing of the experimental curve. Attenuation of the laser resulted in no detectable change in the laser profile.

The possibility of a frequency chirp[13] during the laser pulse, which would greatly alter the nature of the observed effect (see Fig. 1b), was checked both by beating the laser against a cw CO_2 laser and by splitting the pulse and recombining it in an interferometric arrangement with a 12m delay in one arm. The latter technique is sensitive to small frequency changes during the pulse, provided the pulse shape is stable. By careful adjustment of the position of the aperture near the rotating mirror, the frequency chirp could be kept under 2MHz during the pulse

($d\omega/dt \leq 4$MHz/μsec).

Although fluorescent signals were directly observable on an oscilloscope even at our lowest pressure (0.05 torr), signal averaging was necessary in order to obtain accuracy.[14] It is interesting to note that at 0.05 torr, in the region of $\mu\mathcal{E}\tau_p/\hbar = \pi$, the signal originates from approximately 2×10^9 excited molecules. This sensitivity compares well with magnetic resonance.

Experimental data (Fig. 3) reproducibly shows modulation of the fluorescence intensity at a pressure of 0.05 torr. This determines the laser intensity in terms of θ. In order to compare our data with theory, the above analysis must be modified. In CO_2, rotational relaxation is the main T_1 mechanism: once a resonant molecule suffers a collision which changes its rotational state, it no longer interacts significantly with the radiation field. It nevertheless contributes to the measured signal since fluorescence is observed from all rotational levels of the 00°1 vibrational state to the ground state. The proper expression for comparison with our data is thus

$$I_{4.3\mu} \propto \rho_{22}(t_a) + \int_0^{t_a} (\rho_{22}(t)/T_1)\,dt \qquad (6)$$

One effect of this additional term is to cause the modulation to disappear more rapidly as the relaxation times are made shorter than τ_p. Also, the fluorescence increases for shorter T_1 because rotational thermalization tends to restore the population of both interacting levels to their equilibrium values.

An additional complication is the presence of radiation trapping, which introduces a pressure dependence in the relationship between observed fluorescence and excited population.

The data plotted in Fig. 3 are proportional to the intensity of fluorescence immediately after the pulse, normalized to CO_2 pressure and multiplied at each pressure by a factor R(p) introduced to account for radiation trapping. At low pressures, a correction has also been made for a residual linear background signal. Theoretical curves are shown utilizing the known relaxation rate of $1/T_2 = 2 \times 10^7$/torr-sec obtained from pressure broadening studies,[15,16] together with a value of $1/T_1 = 10^7$/torr-sec[17] in Eqs. (2) and (6). The set of numbers R(p) which produce agreement with these theoretical curves (see Fig. 3) is consistent with the trapped lifetimes measured by Kovacs and Javan.[6] However, the uncertainty in this trapping factor for our cell geometry and the insensitivity to T_1 of the curves resulting from Eq. (6) prevent an accurate determination of T_1. Variation of T_2 by 50% produces significantly worse agreement with the set of theoretical curves. The measured τ_p of 0.56 μsec and energy/unit area of 0.024 mJ/cm^2 in the region of the 2π condition give a value of $\mu_{oz}(P20) = (2.0 \pm 0.4) \times 10^{-20}$ esu, in agreement with the value of 1.7×10^{-20} esu deduced from absorption measurements.[15]

The example discussed above shows the potential of the fluorescence method for the measurement of the dipole matrix elements and the relaxation parameters T_1 and T_2 in gases in the presence of inhomogeneous broadening. Photon echoes, self-induced transparency and optical nutation measure similar parameters and thus should be compared with our method. When applicable the fluorescence method has the advantage of simplicity and sensitivity: it can measure relaxation in a sample of total absorption 10^{-5} and relaxation times as short as the pulse length itself. This feature makes the method attractive for the measurement of short

relaxation times, especially using tunable short-pulse lasers.

Despite the advantages claimed above for this technique, we think it necessary to point out certain experimental difficulties. It is necessary to have a stable, reproducible, single mode laser with negligible frequency chirp and uniform amplitude across the beam. It is also important to eliminate any linear background signal arising, for example, from gas adsorbed on cell windows. On the positive side, we note that the magnitude of the dips should be much larger than those reported here if the condition $\tau_p \lesssim \Delta\omega_D^{-1}$ is fulfilled.

Two simplifying assumptions were made in the derivation of Eq. 2: the third level has been neglected, and the radiative reaction on the level pair 1-2 has not been included. It can be shown, using the methods of Feld and Javan,[6] that under the influence of the strong field the radiative lifetime of the level pair 2-3 can change only up to a factor of 2. Superradiant phenomena [18] can also play an important role in determining the shape of the fluorescence curve as well as the fluorescence decay envelope, due to the enhanced rate arising from the superradiant state.[18,1] Both effects are negligible in our experiment.

In conclusion, we note that this technique is also applicable to the study of relaxation in solids and in electronically excited molecular states which are coupled to a quasi continuum of levels.[19] It can be extended to include the time development of the angular distribution and polarization of the emitted radiation, which directly measures the equilibration of the degenerate M-sublevels.

We wish to thank Prof. A. Javan for his continuous encouragement and help in this work. The initial phases of this work were done in cooperation with Dr. C. K. Rhodes, who also bequeathed much of the apparatus.

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5. We know by private communication that many of our colleagues have considered similar experiments: S.R. Hartmann, N.A. Kurnit and I.D. Abella in Ruby; F.T. Arecchi, ourselves and A. Javan, and R.T. Hodgson on the 5401Å line of Ne. A preliminary account of the underlying theory can be found in C.K. Rhodes and A. Szöke, Phys. Rev. 184, 25 (1969) Appendix B, also in the thesis of C.K. Rhodes (M.I.T. 1969 unpublished).
6. Previous spontaneous emission studies have utilized other techniques to obtain related information. See for example: R.H. Cordover, P.A. Bonczyk and A. Javan, Phys. Rev. Letters 18, 730, 1104(E) (1967); H.K. Holt, Phys. Rev. Letters 20, 410 (1968); M.S. Feld and A. Javan, Phys. Rev. 177, 540 (1969);

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7. Let α be the linear absorption coefficient (in cm^{-1}) of a sample whose length is L cm. We define a sample for which $\alpha L \ll 1$ as optically thin. Under these circumstances the x-dependence of the pulse may be neglected.

8. The collisional relaxation problem in gases is discussed in more detail by S.G. Rautian and I.I. Sobelman, Usp. Fiz. Nauk 90, 209 (1966) (Sov. Phys. Uspekhi 9, 701 (1967); B.L. Gyorffy, M. Borenstein and W.E. Lamb, Jr., Phys. Rev. 169, 340 (1968); C.H. Wang and W.J. Tomlinson, Phys. Rev. 181, 116 (1969); E.G. Pestov and S.G. Rautian, JETP 56, 902 (1969) (Sov. Phys. JETP 29, 488 (1969)).

9. In the presence of collisions this description is not general enough. See Ref. 8. Large deviations from our simplified theory are not expected over the range of this experiment.

10. For a more complete derivation see C.K. Rhodes, A. Szöke and A. Javan, Ref. 4; C.K. Rhodes and A. Szöke, Ref. 5.

11. This may be the reason why Hocker and Tang (Ref. 3) have seen the nutation effect. Their theoretical expressions (see also C.L. Tang and H. Statz, Appl. Phys. Letters 10, 145 (1968)) are not equivalent to ours in that they postulate a collision mechanism which strongly couples the degenerate sublevels without loss of phase.

12. This is a characteristic feature of "power broadening" in an inhomogeneously broadened line, where (4) changes to

$$\bar{\rho}_{22}(t_a) = (\rho_{11}^{o}/2)(\mu\mathcal{E}/\hbar)^2 T_1 (\Delta\omega_D/2)^{-1}(1+(\mu\mathcal{E}/\hbar)^2 T_1 T_2)^{-1/2}.$$

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14. For this purpose the availability of a PDP-12 computer greatly facilitated the ease and speed of data taking. The rapidly convergent fast algorithm, $Y_n = Y_{n-1} + (X_n - Y_{n-1})/2^k$; $n/2 \leq 2^k < n$, where X_n is the nth experimental value and Y_n is an approximation to the average of the first n samples, allowed averaging and display of 32 points on each of 2 signals at a 120 cycle rate, so that 2000 samples could be averaged in under 20 sec.

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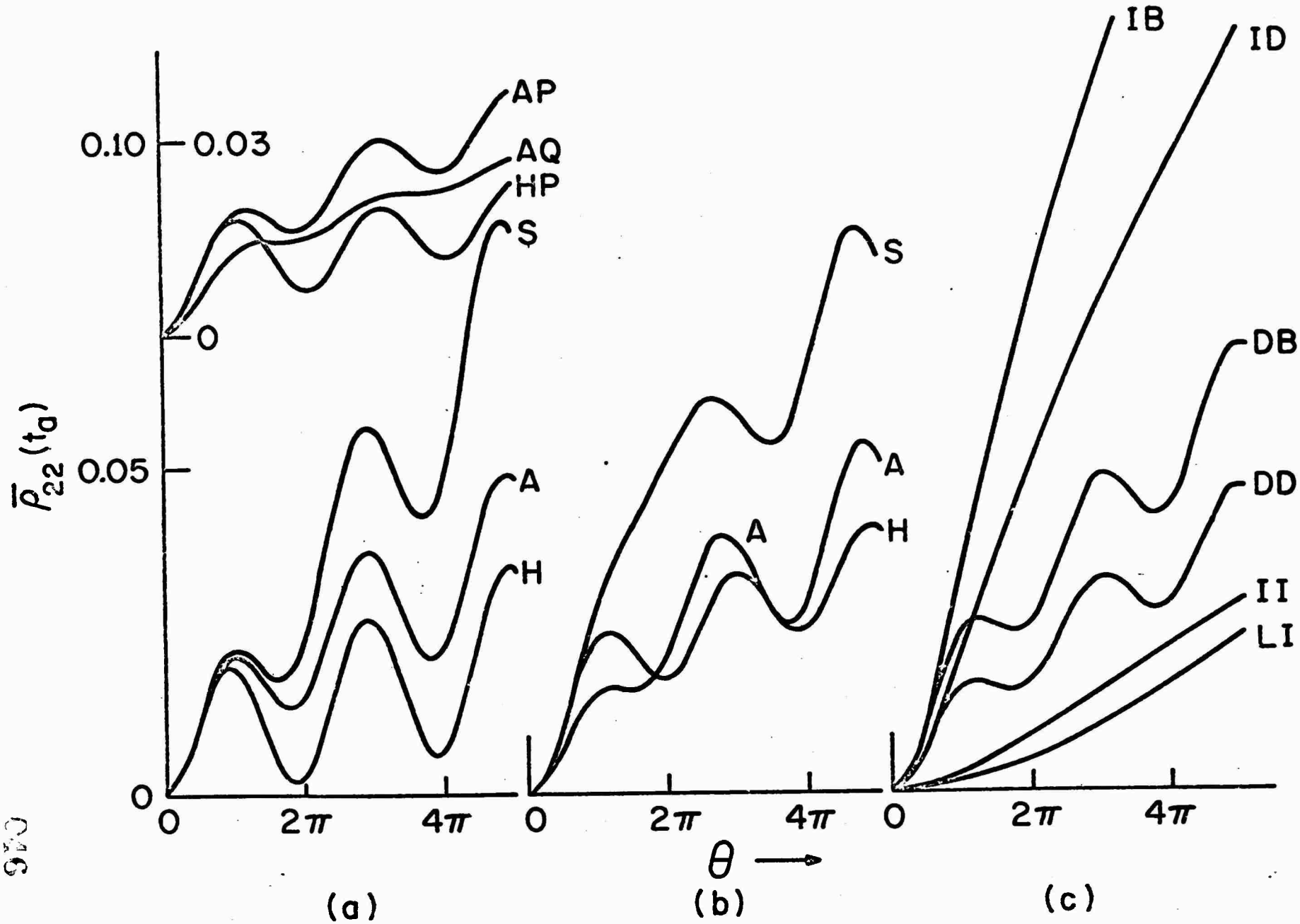
FIGURE CAPTIONS

Fig. 1. Population of upper level as function of θ, calculated for various pulse shapes (S=square, H=approximate hyperbolic-secant, A=approximation of actual pulse of Fig. 2b) with τ_p=0.6 μsec and for $\Delta\omega_D/2\pi$=60 MHz. (a) No relaxation. P,Q indicate average population of degenerate sublevels of high J, $\Delta J=\pm 1$ or $\Delta J=0$ transition. (b) Effect of frequency chirp of 4 MHz/μsec. (c). Effect of relaxation, labeled by T_2,T_1 values (in μsec) as follows: B=∞,D=1, I=0.1, L=0.0125.

Fig. 2. (a) Schematic of experimental apparatus (see text for details). (b) Multiple exposure of laser pulses, 100 nsec/div. (c) Relevant transitions in CO_2.

Fig. 3. Observed fluorescence normalized to CO_2 pressure and multiplied by trapping factor R(p) as function of square root of laser intensity (in units of θ). Theoretical curves from Eq.6, labeled by T_2,T_1 values (in μsec) as follows: C=2, D=1, E=0.5, F=0.25, G=0.2, I=0.1, J=0.05, K=0.025, L=0.0125.Origin is displaced for clarity. Error bars indicate typical uncertainty.


0.10
0.03
0.05
0
$\bar{\rho}_{22}(t_a)$
AP
AQ
HP
S
A
H
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θ
(a)
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IB
ID
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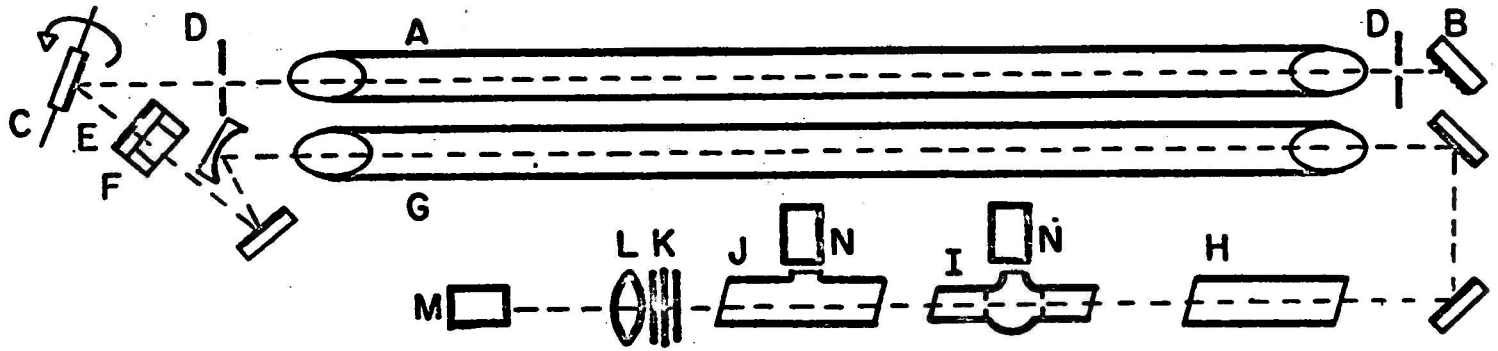
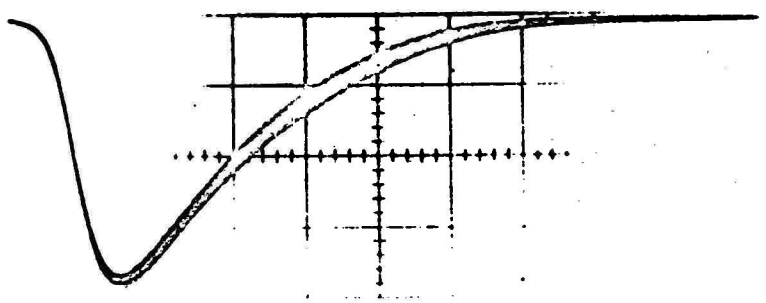
Fig. 1



(a)



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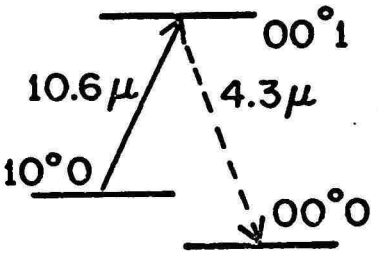

(c)

Fig. 2

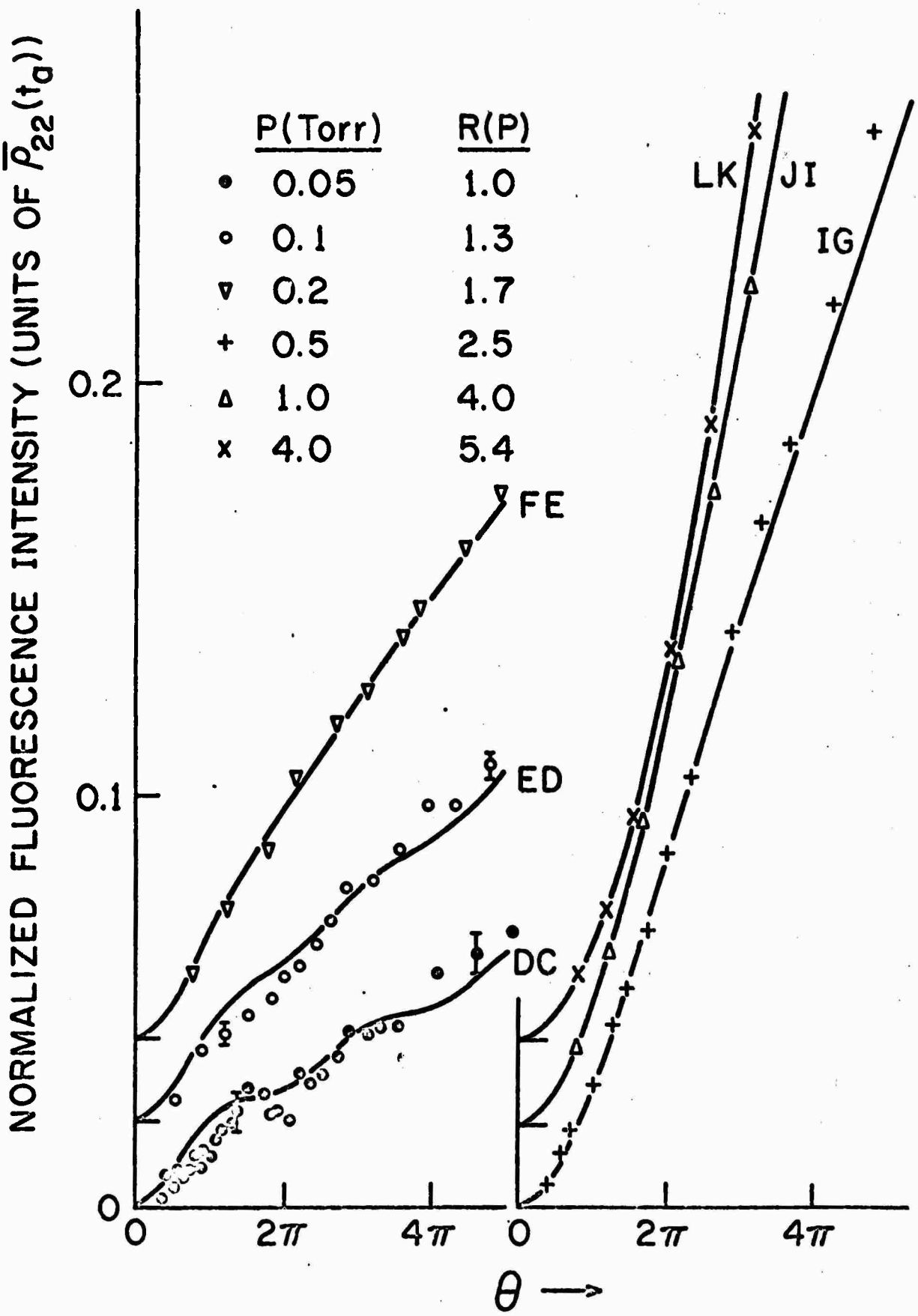
NORMALIZED FLUORESCENCE INTENSITY (UNITS OF $\overline{\rho}_{22}(t_d)$)
P(Torr)
R(P)
0.05
1.0
0.1
1.3
0.2
1.7
0.5
2.5
1.0
4.0
4.0
5.4
FE
ED
DC
LK
JI
IG
0.2
0.1
0
0
2π
4π
0
2π
4π
θ →

Fig. 3

048