AD 665 694

INVESTIGATION OF MOLECULAR GAS LASERS

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February 1968

Massachusetts Institute of Technology
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TITLE

"Investigation of Molecular Gas Lasers"

Semi-Annual Technical Summary Report #1
for Period July 1, 1967 to December 31, 1967

under supervision of

Project Scientist: Professor A. Javan
Telephone: 864-6900, Ext. 5088

under

Contract # N00014-67-A-0204-0014
Project # NR015-717/6-8-67 (case 421)

ARPA Authorization Order #306

Starting Date: July 1, 1967 Expiration Date: June 30, 1968

Amount of Contract $50,000

MIT Project # DSR 70620

This research is part of Project DEFENDER under the joint sponsorship of the Advanced Research Projects Agency, the Office of Naval Research, and the Department of Defense.

Issue Date: February 9, 1968

This progress report consists of a summary and two appendices.

SUMMARY:

The initial work on the study of diffusion and wall deexcitation probability of 001 state in CO_2 is completed. (See Appendix A.) A letter has been prepared and submitted for publication on this subject. The experiment is presently pursued to determine 001 diffusion through He and other buffer gases. Furthermore, preparations are being made to study wall deactivation using wall coating techniques to possibly achieve lifetime enhancement of 001 state.

An additional experiment is underway to study volume quenching probability of the 100 state. (See Appendix B.) The results are currently being analyzed.

A detailed theoretical calculation has been completed on the saturation behavior of a molecular laser amplifier where the time constant of coupling between various rotational levels as well as the various vibrational decay rates play important roles. Details of the "Lamb dip" effect for single-mode single-transition laser oscillation has been related to the important relaxation parameters. Furthermore, mode coupling for a molecular oscillator has been investigated under conditions where the collision broadening is considered to be dominant. It is shown that under appropriate conditions, it is possible to take advantage of mode coupling to achieve

single mode oscillation. Finally, the latter is supported by direct experimental observation. A detailed paper is being prepared for publication of this work.

A detailed theoretical investigation is underway to determine the feasibility of observing transition locking of all the P-branch lines in a self-Q-switched CO_2 laser. Pending the outcome of this investigation, an experiment will probably be undertaken to observe this effect which is expected to yeild ultra-high intensity subnanosecond pulses.

APPENDIX A

Study of Diffusion and Wall Deexcitation Probability of 00^01 State in CO_2

This report summarizes the detailed study of diffusion of excited CO_2 (00^01 state) through pure CO_2 and measurement of its wall deexcitation probability. A Q-switched CO_2 laser pulse incident on a separate cell containing pure CO_2 increases suddenly the 00^01 level population; the subsequent decay is monitored via decay of the spontaneous 4.3μ radiation arising from the 00^01 - 00^00 transition. The measurements are made at room temperature with pressures from 1 - 10000μ - Hg.

The decay rate due to diffusion alone does not exhibit an inverse (pressure X radius^2) dependence. This departure allows one to calculate the wall deexcitation probability. The measured coefficient for the 00^01 state differs from the self-diffusion coefficient obtained from viscosity measurements which reflect primarily ground state behavior. Energy transfer effects in 00^01 - 00^00 type collisions may be responsible for the difference.

As in earlier experiments[(1)] the test cell is mounted within the cavity of a Q-switched CO_2 laser. Pyrex, brass, mylar and teflon cylinders with diameters, 2.54, 1.50, 1.20, and 0.88 cm are placed in this cell. Each insert has a small infrared transmitting window.[(2)] The spontaneous emission is detected with a Ge:Au (77^0K) element whose output is amplified and stored by a multi-channel signal averager. Approximately 30 data points are taken for each cell radius and surface. A plot of averaged signal vs. time yields in all cases a single exponential. Signals are observable at pressures as low as one μ - Hg.

Figure 1 gives decay rate vs. pressure data and the best fit obtained from theory below. Figure 2 displays the best theoretical fit to the decay rate vs. $1/(\text{radius})^2$ at various pressures; at low pressures the deviation from a straight line is apparent.

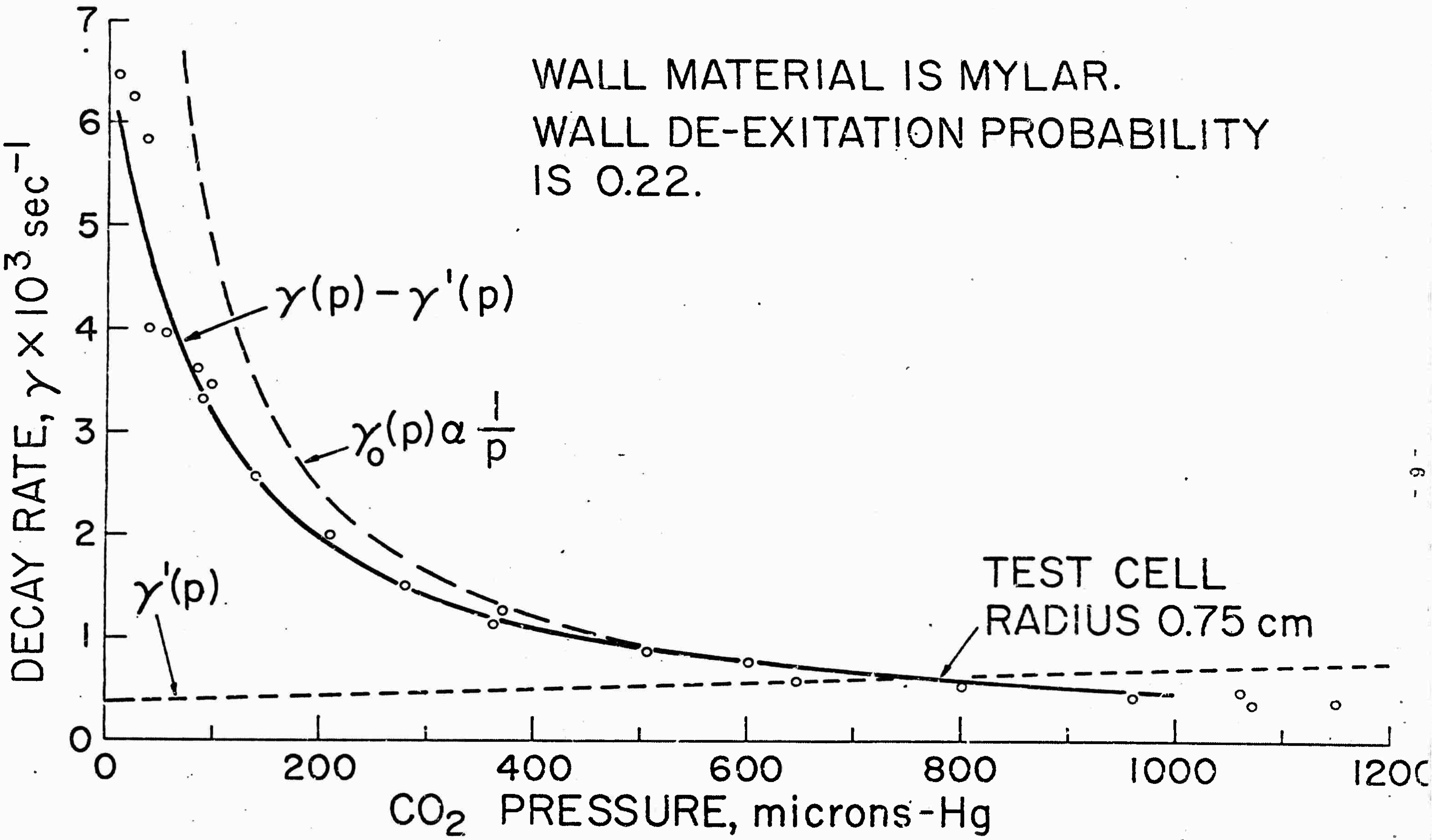
A diffusion equation with a volume quenching term $(-\rho_{exc.}\,\gamma_{vol.\ quenching})$, where $\rho_{exc.}$ is density of excited CO_2 and $\gamma_{vol.\ quenching}$ is the volume quenching decay rate, and a boundary condition describing partially reflecting walls gives the pressure dependent decay rate. The continuity equation

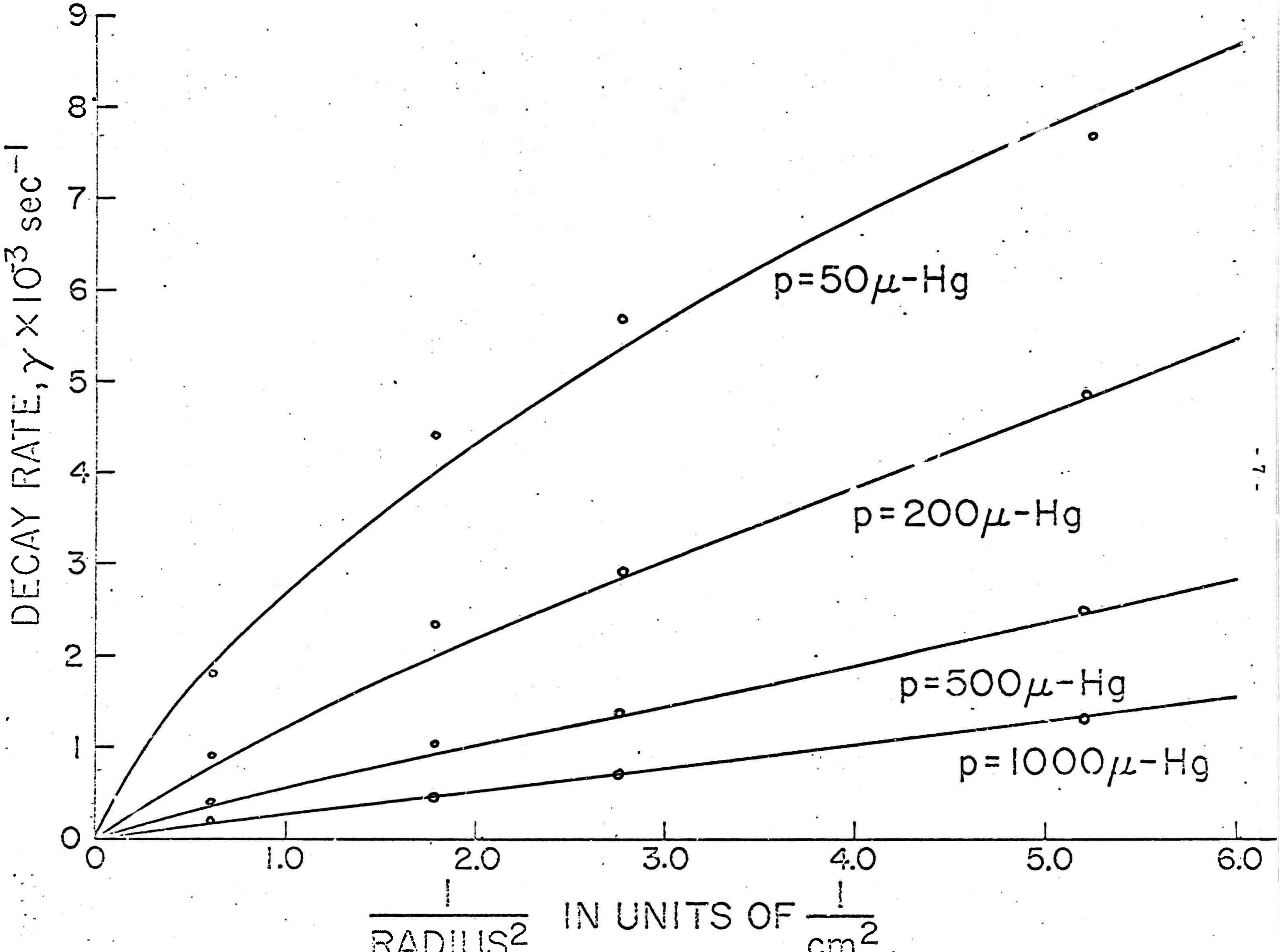
$$\int_{surface} \text{flux} \cdot dS = - \int_{volume} \dot{\rho}_{exc.}\, dv$$

gives the boundary condition relating the density and its first derivative at the wall; the outward flux at the wall is $(1-\beta)\left(\rho_{exc.}\frac{\bar{v}}{4} - \frac{D}{2}\frac{\partial \rho_{exc.}}{\partial x}\right)\Big|_{wall}$ where $\bar{v}$, is the mean speed, D, the diffusion coefficient, and β is the wall reflection probability.(3) In cylindrical coordinates, the lowest order solution is $\rho\,(r,t) = J_o\,(\mu \frac{r}{r_o})\, e^{-\gamma t}$ where $\gamma = \frac{\mu^2 D}{r_o^2} + \gamma_{vol.\ quenching}$, μ is the solution to the boundary equation, and r_o, the test cell radius. The decay rate $\gamma_{vol.\ quenching}$ is linear in pressure and is small at pressures appreciably below one torr. (See Figure 1) Although D is inversely pressure dependent, γ does not exhibit a 1/pressure dependence throughout the range 10 - 1000μ - Hg, since the boundary equation makes μ^2 dependent on pressure and radius. At low pressure μ^2 is linear in pressure and approaches a constant value.[4] In the region between 10 - 700μ- Hg, μ^2 is a function of pressure and the diffusion decay rate does not have a 1/pressure dependence; only near one torr is μ^2 a constant and behaves as 1/pressure.

The measured diffusion coefficient is .07 $\pm$ .01 cm^2 - sec^{-1} which gives the diffusion cross section 9.1 $\pm$ 1 x 10^{-15} cm^2. The self-diffusion constant and cross-section obtained from viscosity measurements are 0.11 cm^2 - sec^{-1} and 5.3 x 10^{-15} cm^2 respectively.[5] The wall deexcitation probability, $(1 - \beta)$, is 0.22 $\pm$ .08 for the four surfaces studied; it is quite probable that the surfaces used have similar surface contaminants, such as absorbed CO_2.

This present experiment improves the value for the previously obtained 00^01 state volume quenching rate;[1, 6] it is now measured as 335 $\pm$ 5 sec^{-1} (mm. - Hg.)$^{-1}$.


WALL MATERIAL IS MYLAR.
WALL DE-EXITATION PROBABILITY
IS 0.22.
γ(p) − γ'(p)
γ₀(p) α 1/p
γ'(p)
TEST CELL
RADIUS 0.75 cm
DECAY RATE, γ × 10³ sec⁻¹
CO2 PRESSURE, microns-Hg
0
1
2
3
4
5
6
7
0
200
400
600
800
1000
1200



DECAY RATE, γ × 10³ sec⁻¹
0
1
2
3
4
5
6
7
8
9
p=50μ-Hg
p=200μ-Hg
p=500μ-Hg
p=1000μ-Hg
0
1.0
2.0
3.0
4.0
5.0
6.0
1/RADIUS² IN UNITS OF 1/cm²

APPENDIX B

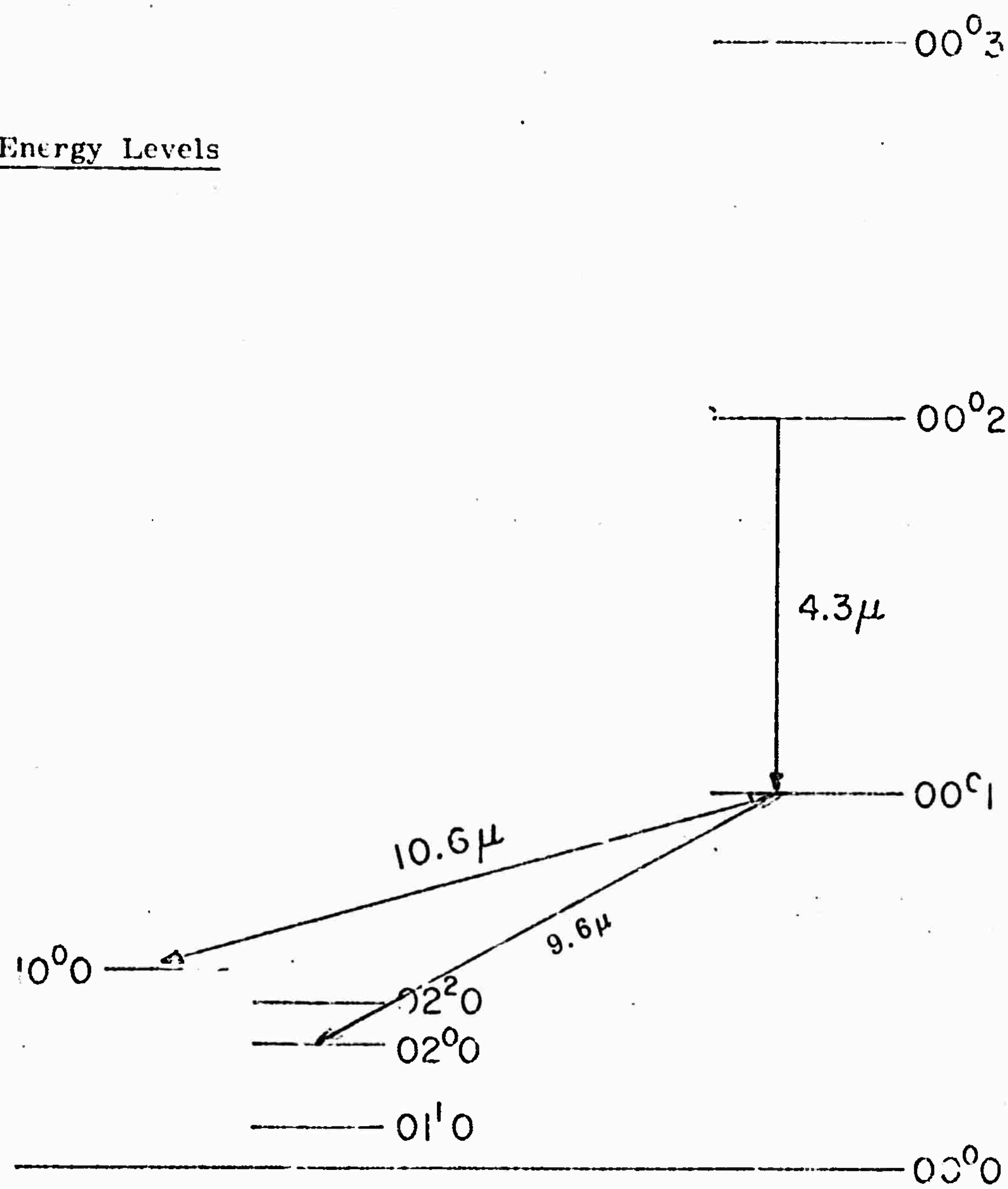
CO_2 Relaxation Studies

Efforts have been made to examine the collisional decay modes of the 10^00 vibrational level in the CO_2 molecule. Since information is available on the relaxation rates of the 00^01 level,[1] it is possible to infer the behavior of the 10^00 level by studying the $00^01 \leftrightarrow 10^00$ transition at 10.6μ. For numerous reasons, it is important to study this process in the absence of a gaseous discharge where the equilibrium population distribution is governed by thermal processes. This is the approach used: Essentially, we consider the three level system composed of the 10^00, 02^00 Fermi resonance pair and the 00^01 level. Figure (1) illustrates these levels. For discussion, we designate the thermal population densities of the 00^01, 10^00, and 02^00 levels as n_1, n_2, and n_3 respectively. These population densities determine the linear susceptibility. Hence, the linear susceptibility of the medium at or near 10.6μ is proportional to the factor $(n_2 - n_1)$.

$$\chi_{(10.6)} \propto (n_2 - n_1) \qquad (1)$$

The experiment incorporates the following sequence of events. The CO_2 gas is initially at equilibrium with certain equilibrium population densities, denoted by n_1^o, n_2^o, and n_3^o. Then at time $t = t_o$, the medium is subjected to a saturating optical pulse at 9.6μ obtained from a separate Q-switch laser

(1) L. Hocker, M. Kovacs, C. Rhodes, G. Flynn, and A. Javan, Phys. Rev. Letters 17, 5, 233 (1966).



CO$_2$ Energy Levels

FIGURE 1

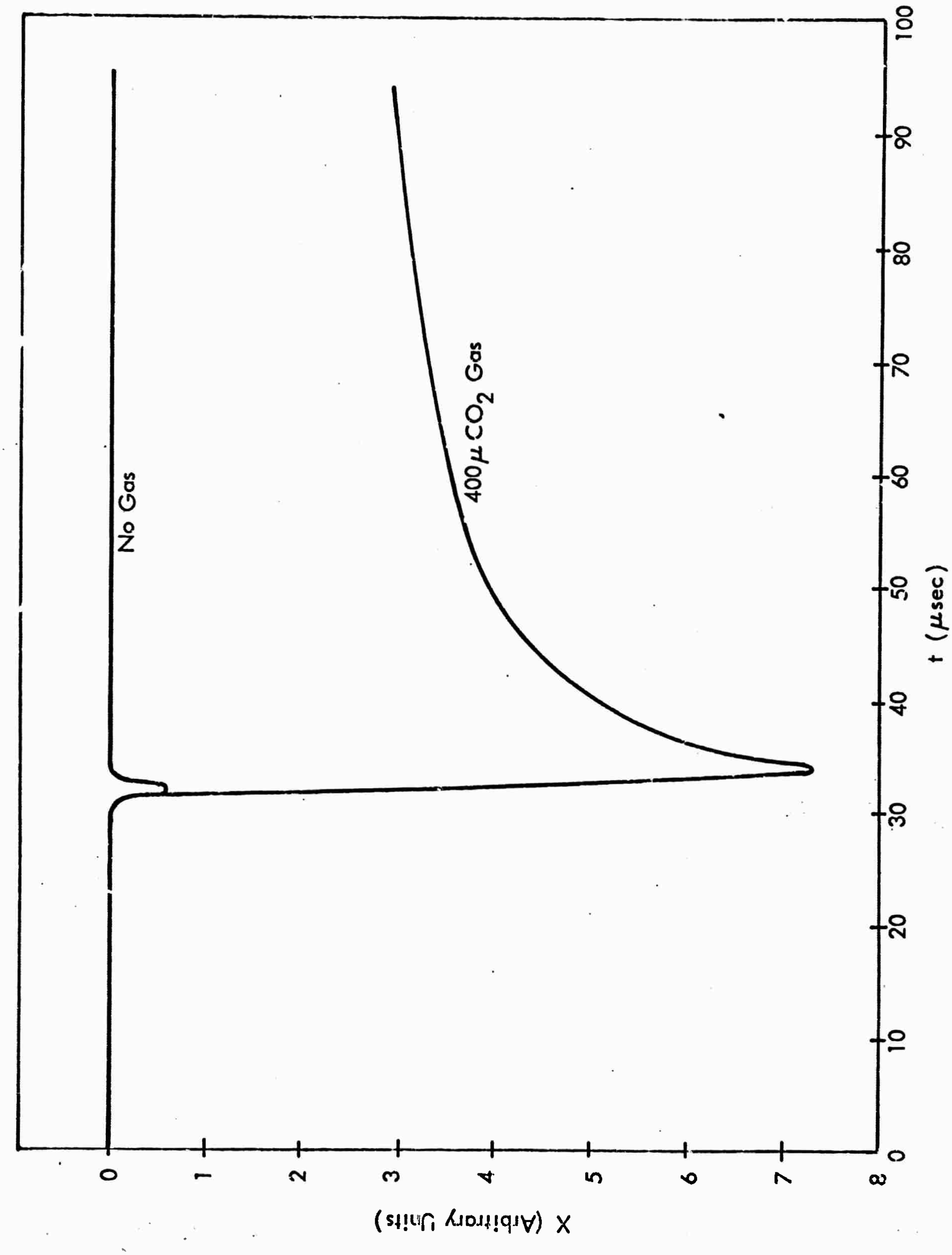


Fig. 2a CO_2 Relaxation Data for 400 μ Hg

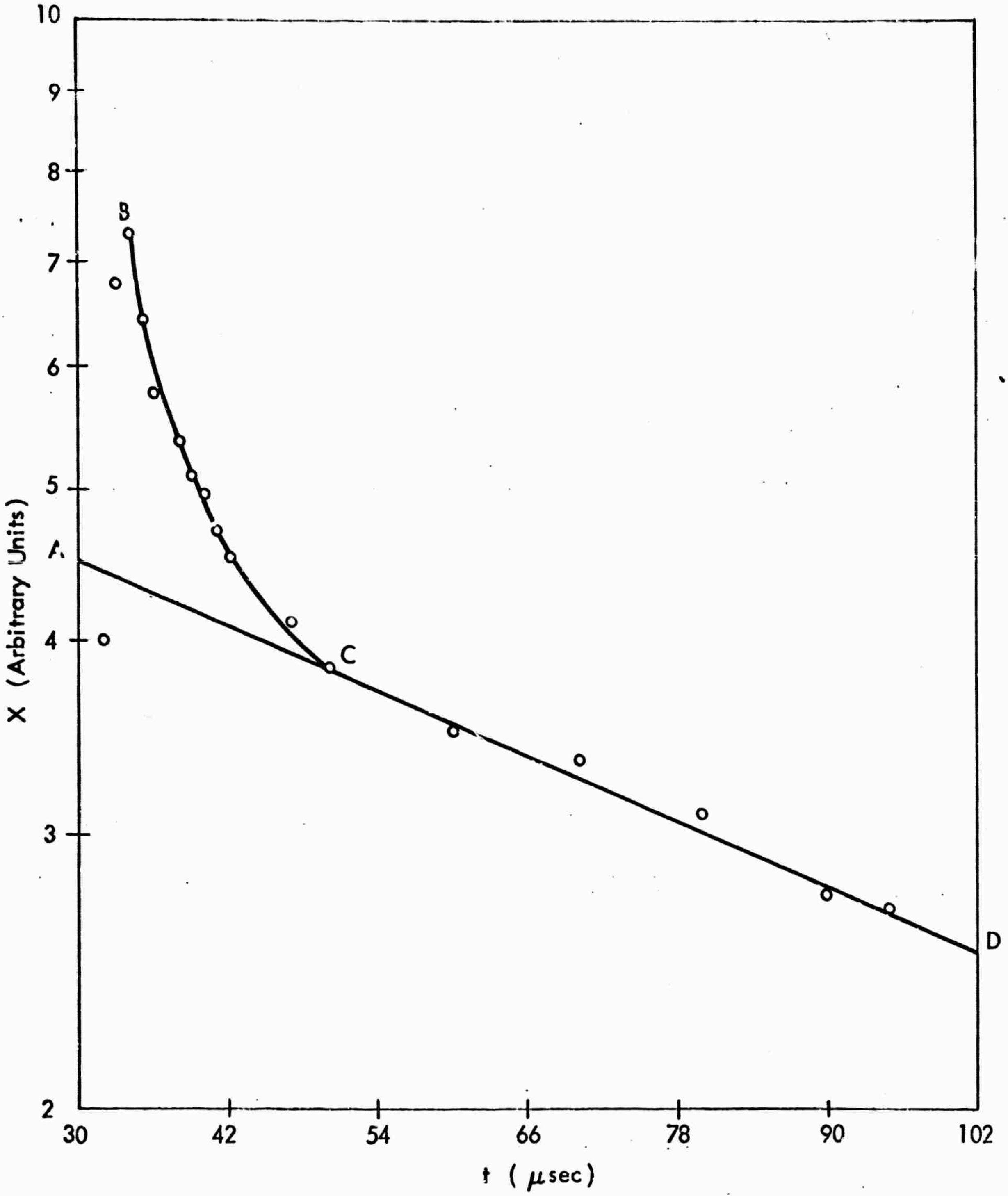


Fig. 2b CO_2 Relaxation Data for 400 μ Hg

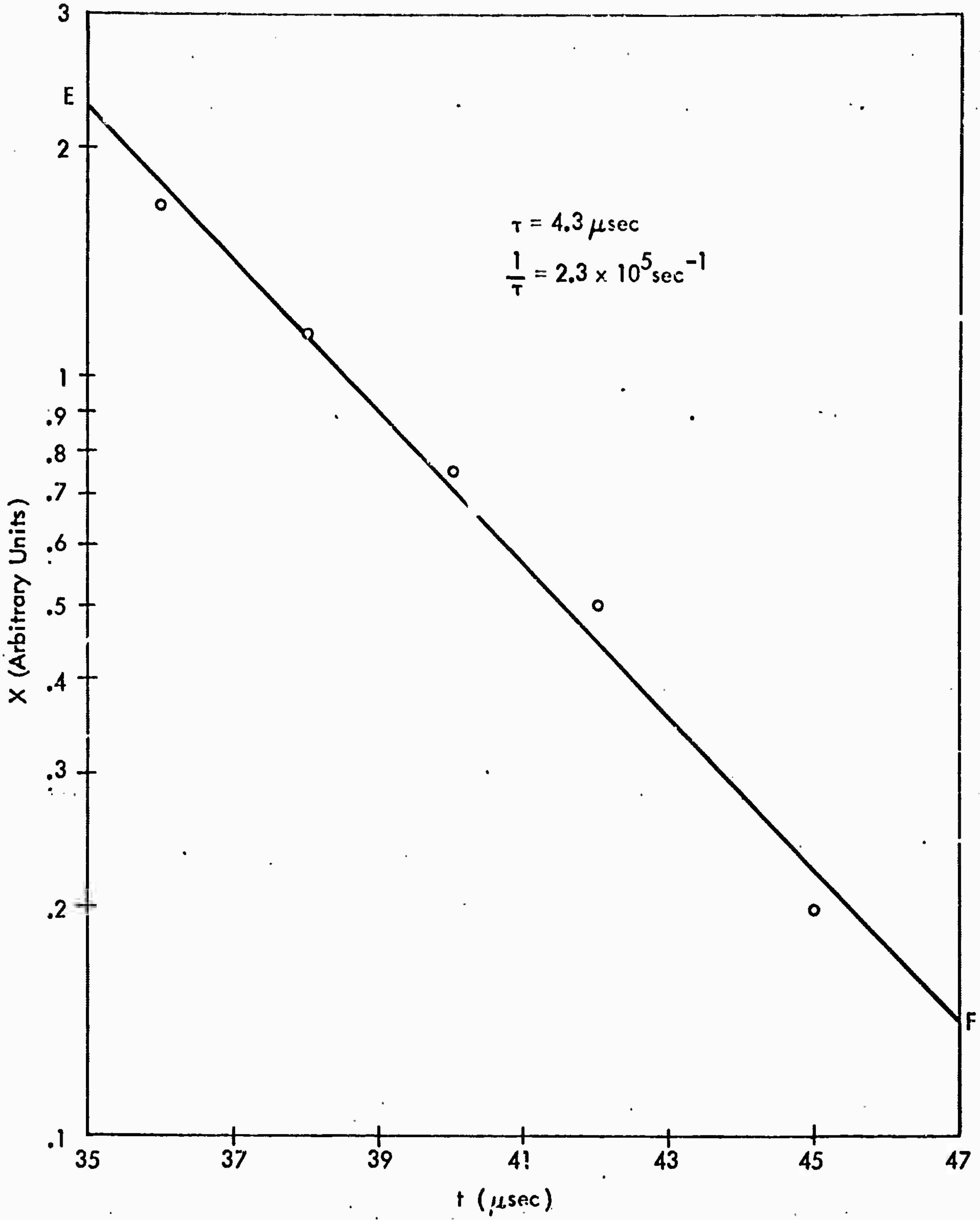


Fig. 2c CO_2 Relaxation Data for 400 μ Hg pulse. This pulse momentarily equates the population densities n_1 and n_3. The pulse length is assumed to be short in comparison with all the relevant relaxation times. Thus at $t = t_o + \delta$, where δ is the effective 9.6μ pulse length, the non-equilibrium population densities are the following.

$$n_1 \Big|_{t_o+} = n_3 \Big|_{t_o+} = \frac{n_1^o + n_3^o}{2} \tag{2}$$

$$n_2 \Big|_{t_o+} = n_2^o \tag{3}$$

As time passes collisions among the gas molecules reestablish the original equilibrium. This evolution will cause the linear susceptibility $\chi_{(10.6)}$ to vary through the factor $(n_{2(t)} - n_{1(t)})$. This quantity is measured directly by monitoring the absorption of the gas sample at 10.6μ before and after the 9.6μ saturating pulse.

Typical experimental results are shown in Figures (2a), (2b) and (2c). The ordinate X is proportional to $\chi_{(10.6)}$ in arbitrary units. The raw data is illustrated in Figure (2a). Figures (2b) and (2c) are semi-log plots of the same data which are designed to project the exponential decay properties of the original curve (2a).

Unclassified
Security Classification

DOCUMENT CONTROL DATA - R&D

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1. ORIGINATING ACTIVITY (Corporate author) Massachusetts Institute of Technology Cambridge, Massachusetts	2a. REPORT SECURITY CLASSIFICATION Unclassified 2b GROUP

3. REPORT TITLE

"Investigation of Molecular Gas Lasers"

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)
Semi-Annual Technical Summary Report #1 7/1/67 - 12/31/67

5. AUTHOR(S) (Last name, first name, initial)

Javan, Ali - Professor | Kelly, Michael J.
Kovacs, Mark A. | Rhodes, Charles K.

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
February 1968	13	1

8a. CONTRACT OR GRANT NO. N00014-67-A-0204-0014 b. PROJECT NO. NR-015-717/6-8-67 (Code 421) c. ARPA Authorization # 306 d	9a. ORIGINATOR'S REPORT NUMBER(S) Semi-Annual Technical Report #1 9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)

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13. ABSTRACT

The diffusion coefficient for 001 state of CO_2 into CO_2 is measured and is shown to be twice larger than the self-diffusion coefficient of CO_2. The wall deexcitation 001 state is also determined in detail. In another experiment, the collision lifetime of 100 state, the lower laser level is measured for the first time and studied in detail. A theoretical calculation is completed dealing with details of a CO_2 amplifier saturation and manifestation of "Lamb dip" effect.

DD FORM 1 JAN 64 1473

Unclassified
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	ROLE	WT	ROLE	WT	ROLE	WT
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high power gas lasers						
molecular laser amplifier						

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