

UNCLASSIFIED

AD 665 694

INVESTIGATION OF MOLECULAR GAS LASERS

Ali Javan, et al

Massachusetts Institute of Technology
Cambridge, Massachusetts

February 1968

Processed for . . .

DEFENSE DOCUMENTATION CENTER
DEFENSE SUPPLY AGENCY



U. S. DEPARTMENT OF COMMERCE / NATIONAL BUREAU OF STANDARDS / INSTITUTE FOR APPLIED TECHNOLOGY

UNCLASSIFIED

Massachusetts Institute of Technology
Cambridge, Massachusetts 02139

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-C014
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 70520

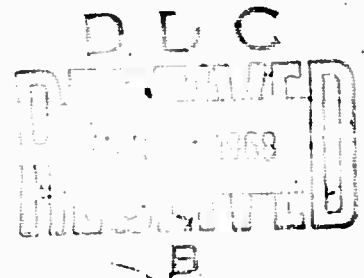
This research is part of Project DEFENDER under the joint sponsorship of the Advanced Research Projects Agency, the Office of Naval Research, and the Department of Defense.

Reproduction in whole or in part is permitted for any purpose of the United States Government.

Distribution of this document is unlimited.

Issue Date: February 9, 1968

Reproduced by the
CLEARINGHOUSE
for Federal Scientific & Technical
Information Springfield Va. 22151



AD 665694

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 CO₂ laser. Pending the outcome of this investigation, an experiment will probably be undertaken to observe this effect which is expected to yield ultra-high intensity subnanosecond pulses.

APPENDIX A

Study of Diffusion and Wall Deexcitation Probability of 00^0_1 State in CO_2

This report summarizes the detailed study of diffusion of excited CO_2 (00^0_1 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 00^0_1 level population; the subsequent decay is monitored via decay of the spontaneous 4.3μ radiation arising from the $00^0_1 - 00^0_0$ transition. The measurements are made at room temperature with pressures from 1 - 10000 μ - Hg.

The decay rate due to diffusion alone does not exhibit an inverse (pressure X radius²) dependence. This departure allows one to calculate the wall deexcitation probability. The measured coefficient for the 00^0_1 state differs from the self-diffusion coefficient obtained from viscosity measurements which reflect primarily ground state behavior. Energy transfer effects in $00^0_1 - 00^0_0$ 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°K) 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 ($-\rho_{\text{exc.}} \gamma_{\text{vol. quenching}}$), where $\rho_{\text{exc.}}$ is density of excited CO_2 and $\gamma_{\text{vol. quenching}}$ is the volume quenching decay rate, and a boundary condition describing partially reflecting walls gives the pressure dependent decay rate. The continuity equation

$$\int_{\text{surface}} \text{flux} \cdot dS = - \int_{\text{volume}} \dot{\rho}_{\text{exc.}} dv \text{ gives the boundary condition relating the}$$

density and its first derivative at the wall; the outward flux at the wall is

$$(1 - \beta) \left(\rho_{\text{exc.}} \frac{\bar{v}}{4} - \frac{D}{2} \frac{\partial \rho_{\text{exc.}}}{\partial x} \right) \Big|_{\text{wall}} \text{ where } \bar{v}, \text{ is the mean speed, } D, \text{ the diffusion}$$

coefficient, and β is the wall reflection probability. ⁽³⁾ In cylindrical coordi-

nates, the lowest order solution is $\rho(r, t) = J_0\left(\mu \frac{r}{r_0}\right) e^{-\gamma t}$ where

$$\gamma = \frac{\mu^2 D}{r_0^2} + \gamma_{\text{vol. quenching}}, \mu \text{ is the solution to the boundary equation, and } r_0,$$

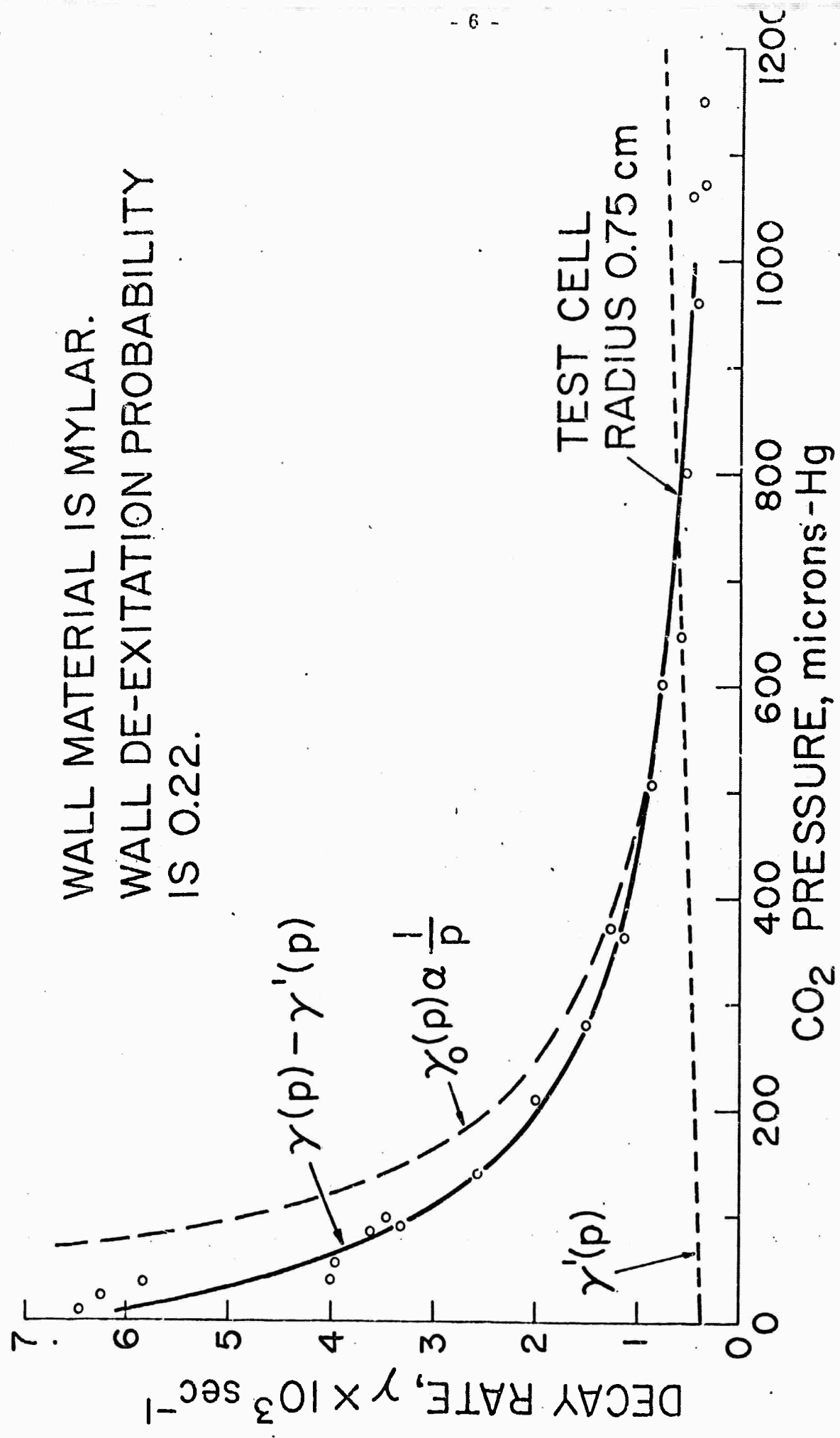
the test cell radius. The decay rate $\gamma_{\text{vol. quenching}}$ is linear in pressure and is small at pressures appreciably below one torr. (See Figure 1) Although D is inversely pressure dependent, γ does not exhibit a $1/\text{pressure}$ dependence throughout the range $10 - 1000 \mu$ - Hg, since the boundary equation makes μ^2 dependent on pressure and radius. At low pressure μ^2 is linear in pressure

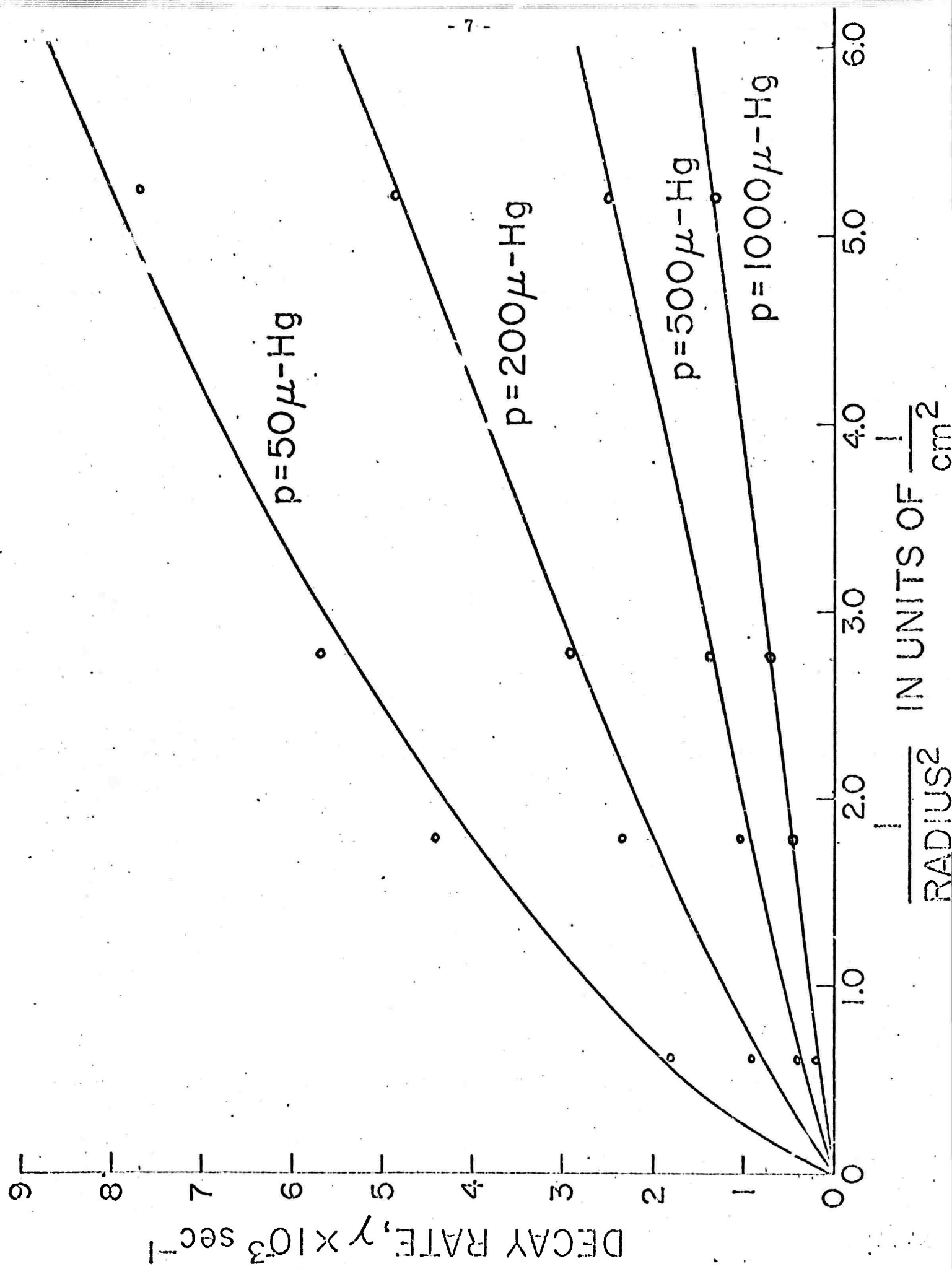
and approaches a constant value. ⁽⁴⁾ In the region between 10 - 700 μ -Hg, μ^2 is a function of pressure and the diffusion decay rate does not have a 1/pressure dependence; only near one torr is μ^2 a constant and behaves as 1/pressure.

The measured diffusion coefficient is $.07 \pm .01 \text{ cm}^2 - \text{sec}^{-1}$ which gives the diffusion cross section $9.1 \pm 1 \times 10^{-15} \text{ cm}^2$. The self-diffusion constant and cross-section obtained from viscosity measurements are $0.11 \text{ cm}^2 - \text{sec}^{-1}$ and $5.3 \times 10^{-15} \text{ cm}^2$ 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 CO^0_1 state volume quenching rate; ^(1, 6) it is now measured as $335 \pm 5 \text{ sec}^{-1} (\text{mm.} - \text{Hg.})^{-1}$.

WALL MATERIAL IS MYLAR.
WALL DE-EXCITATION PROBABILITY
IS 0.22.





APPENDIX B

CO₂ Relaxation Studies

Efforts have been made to examine the collisional decay modes of the $10^0 0$ vibrational level in the CO₂ molecule. Since information is available on the relaxation rates of the $00^0 1$ level, ⁽¹⁾ it is possible to infer the behavior of the $10^0 0$ level by studying the $00^0 1 \leftrightarrow 10^0 0$ 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 $10^0 0$, $02^0 0$ Fermi resonance pair and the $00^0 1$ level. Figure (1) illustrates these levels. For discussion, we designate the thermal population densities of the $00^0 1$, $10^0 0$, and $02^0 0$ 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 $(n_2 - n_1)$.

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

The experiment incorporates the following sequence of events. The CO₂ gas is initially at equilibrium with certain equilibrium population densities, denoted by n_1^0 , n_2^0 , and n_3^0 . Then at time $t = t_0$, the medium is subjected to a saturating optical pulse at 9.6μ obtained from a separate Q-switch laser

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

CO₂ Energy Levels

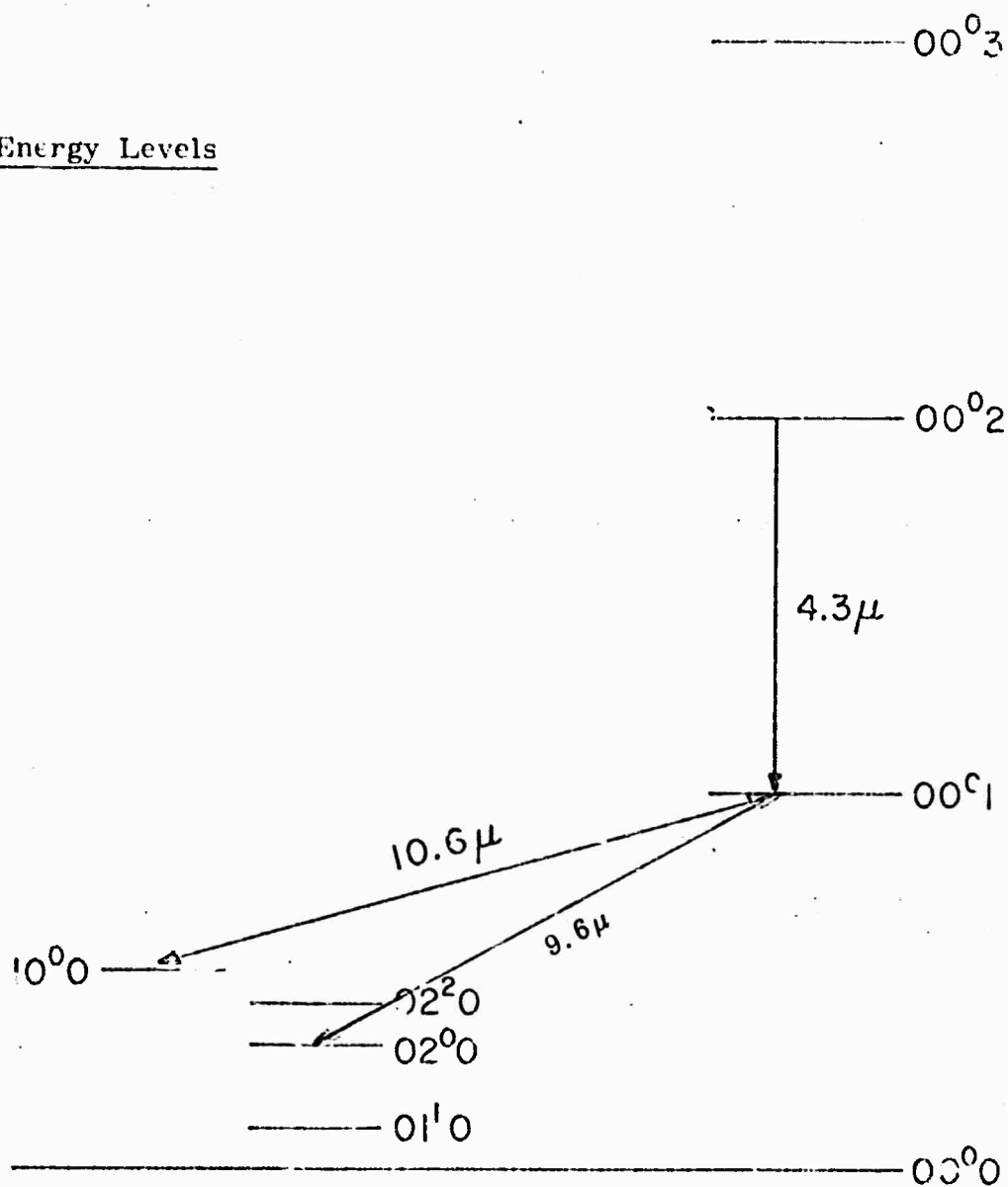


FIGURE 1

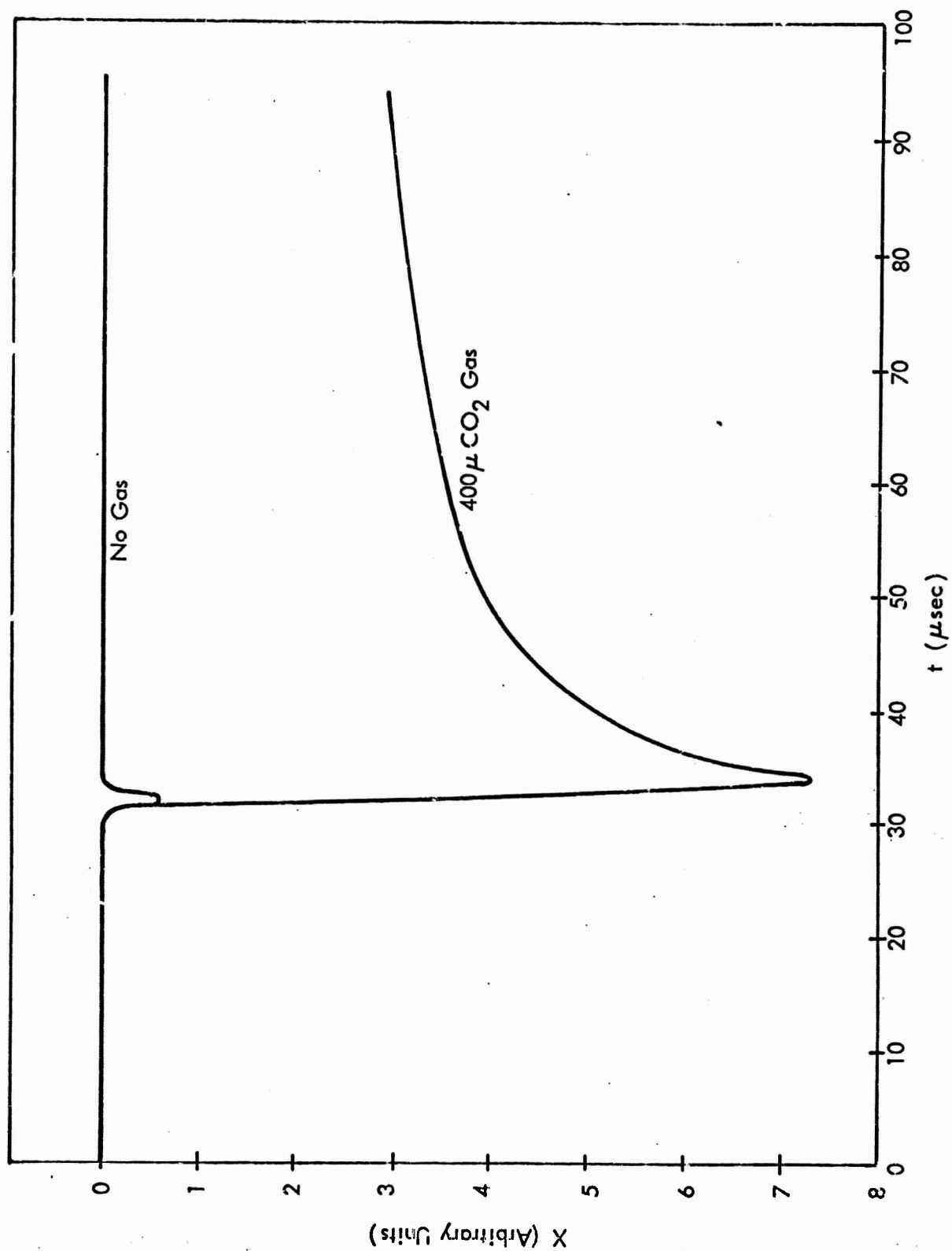


Fig. 2a CO₂ Relaxation Data for 400 μ Hg

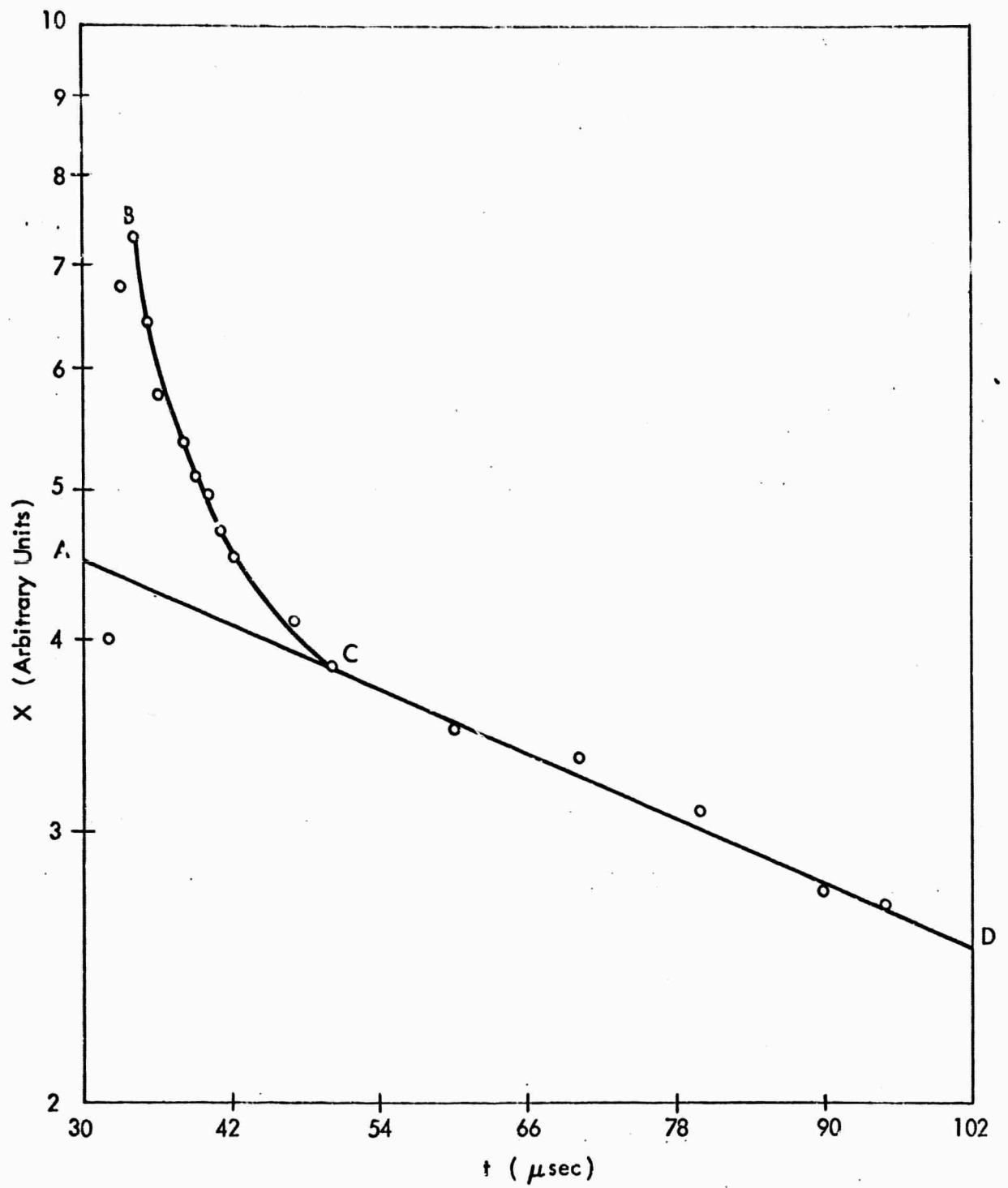


Fig. 2b CO₂ Relaxation Data for 400 μ Hg

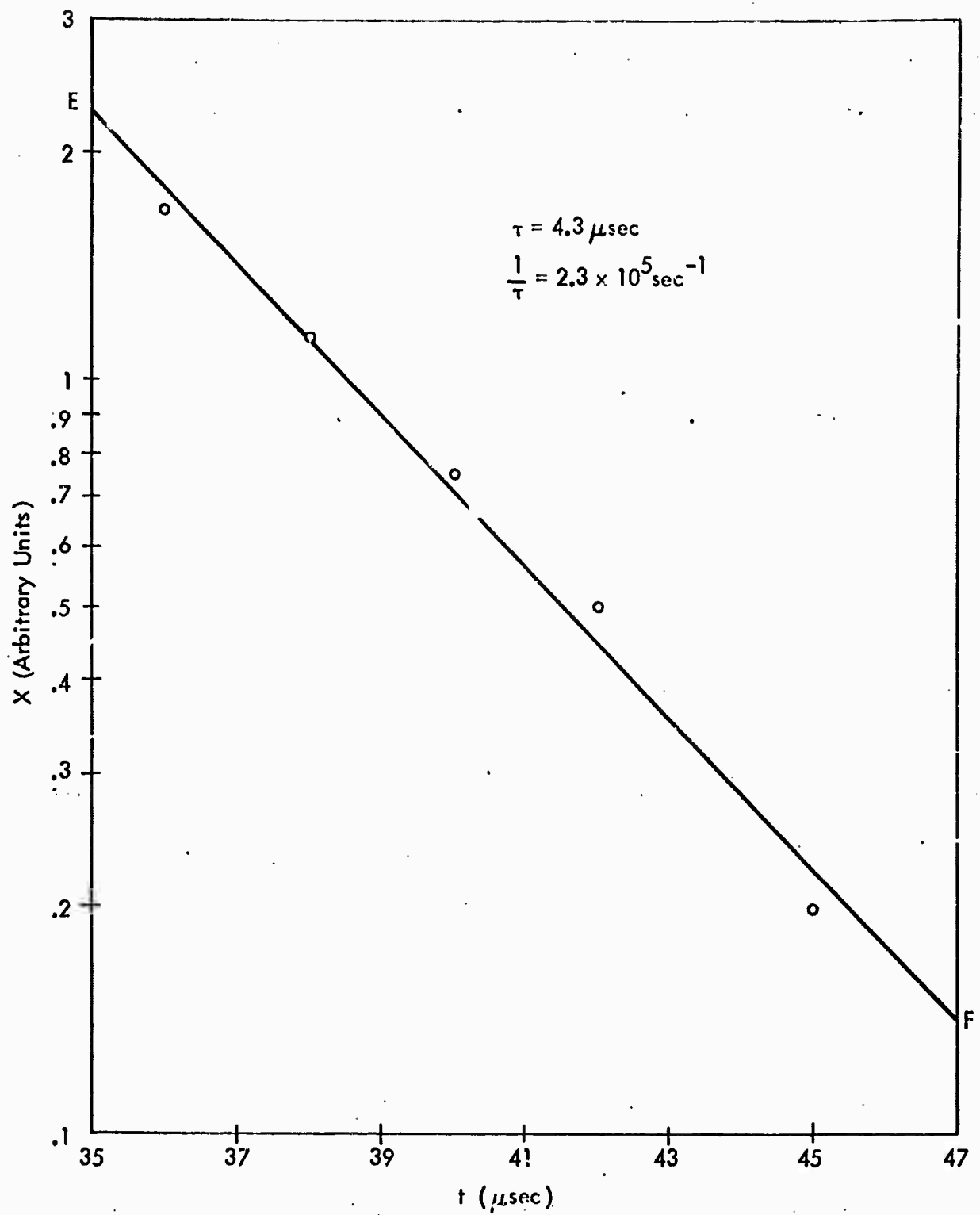


Fig. 2c CO₂ Relaxation Data for 400 μ Hg

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 $t = t_0 + \delta$, where δ is the effective 9.6μ pulse length, the non-equilibrium population densities are the following.

$$n_1 \Big|_{t_0+} = n_3 \Big|_{t_0+} = \frac{n_1^0 + n_3^0}{2} \quad (2)$$

$$n_2 \Big|_{t_0+} = n_2^0 \quad (3)$$

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

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

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 Kovacs, Mark A. Kelly, Michael J. Rhodes, Charles K.			
6. REPORT DATE February 1968		7a. TOTAL NO. OF PAGES 13	7b. NO. OF REFS 1
8a. CONTRACT OR GRANT NO. N00014-67-A-0204-0014		9a. ORIGINATOR'S REPORT NUMBER(S) Semi-Annual Technical Report #1	
b. PROJECT NO. NR-015-717/6-8-67 (Code 421)			
c. ARPA Authorization # 306		9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)	
10. AVAILABILITY/LIMITATION NOTICES Distribution of this document is unlimited.			
11. SUPPLEMENTARY NOTES		12. SPONSORING MILITARY ACTIVITY Office of Naval Research Advanced Research Projects Agency	
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

Security Classification

14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
molecular relaxation high power gas lasers molecular laser amplifier						

INSTRUCTIONS

1. ORIGINATING ACTIVITY: Enter the name and address of the contractor, subcontractor, grantee, Department of Defense activity or other organization (corporate author) issuing the report.

2a. REPORT SECURITY CLASSIFICATION: Enter the overall security classification of the report. Indicate whether "Restricted Data" is included. Marking is to be in accordance with appropriate security regulations.

2b. GROUP: Automatic downgrading is specified in DoD Directive 5200.10 and Armed Forces Industrial Manual. Enter the group number. Also, when applicable, show that optional markings have been used for Group 3 and Group 4 as authorized.

3. REPORT TITLE: Enter the complete report title in all capital letters. Titles in all cases should be unclassified. If a meaningful title cannot be selected without classification, show title classification in all capitals in parentheses immediately following the title.

4. DESCRIPTIVE NOTES: If appropriate, enter the type of report, e.g., interim, progress, summary, annual, or final. Give the inclusive dates when a specific reporting period is covered.

5. AUTHOR(S): Enter the name(s) of author(s) as shown on or in the report. Enter last name, first name, middle initial. If military, show rank and branch of service. The name of the principal author is an absolute minimum requirement.

6. REPORT DATE: Enter the date of the report as day, month, year, or month, year. If more than one date appears on the report, use date of publication.

7a. TOTAL NUMBER OF PAGES: The total page count should follow normal pagination procedures, i.e., enter the number of pages containing information.

7b. NUMBER OF REFERENCES: Enter the total number of references cited in the report.

8a. CONTRACT OR GRANT NUMBER: If appropriate, enter the applicable number of the contract or grant under which the report was written.

8b, 8c, & 8d. PROJECT NUMBER: Enter the appropriate military department identification, such as project number, subproject number, system numbers, task number, etc.

9a. ORIGINATOR'S REPORT NUMBER(S): Enter the official report number by which the document will be identified and controlled by the originating activity. This number must be unique to this report.

9b. OTHER REPORT NUMBER(S): If the report has been assigned any other report numbers (either by the originator or by the sponsor), also enter this number(s).

10. AVAILABILITY/LIMITATION NOTICES: Enter any limitations on further dissemination of the report, other than those imposed by security classification, using standard statements such as:

- (1) "Qualified requesters may obtain copies of this report from DDC."
- (2) "Foreign announcement and dissemination of this report by DDC is not authorized."
- (3) "U. S. Government agencies may obtain copies of this report directly from DDC. Other qualified DDC users shall request through _____."
- (4) "U. S. military agencies may obtain copies of this report directly from DDC. Other qualified users shall request through _____."
- (5) "All distribution of this report is controlled. Qualified DDC users shall request through _____."

If the report has been furnished to the Office of Technical Services, Department of Commerce, for sale to the public, indicate this fact and enter the price, if known.

11. SUPPLEMENTARY NOTES: Use for additional explanatory notes.

12. SPONSORING MILITARY ACTIVITY: Enter the name of the departmental project office or laboratory sponsoring (paying for) the research and development. Include address.

13. ABSTRACT: Enter an abstract giving a brief and factual summary of the document indicative of the report, even though it may also appear elsewhere in the body of the technical report. If additional space is required, a continuation sheet shall be attached.

It is highly desirable that the abstract of classified reports be unclassified. Each paragraph of the abstract shall end with an indication of the military security classification of the information in the paragraph, represented as (TS), (S), (C), or (U).

There is no limitation on the length of the abstract. However, the suggested length is from 150 to 225 words.

14. KEY WORDS: Key words are technically meaningful terms or short phrases that characterize a report and may be used as index entries for cataloging the report. Key words must be selected so that no security classification is required. Identifiers, such as equipment model designation, trade name, military project code name, geographic location, may be used as key words but will be followed by an indication of technical context. The assignment of links, rules, and weights is optional.