INVESTIGATION OF MOLECULAR GAS LASERS

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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 CO₂ 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 yield ultra-high intensity subnanosecond pulses.
Study of Diffusion and Wall Deexcitation Probability of $00^1 0^1$ State in $CO_2$

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

The decay rate due to diffusion alone does not exhibit an inverse ($pressure \times radius^2$) dependence. This departure allows one to calculate the wall deexcitation probability. The measured coefficient for the $00^1 0^1$ state differs from the self-diffusion coefficient obtained from viscosity measurements which reflect primarily ground state behavior. Energy transfer effects in $00^1 0^1 - 00^0 0^0$ 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 $\text{mm}^\text{Hg}$.

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

$$\int \text{flux} \cdot dS = - \int \rho_{\text{exc.}} \cdot dV$$

where $\bar{v}$, is the mean speed, $D$, the diffusion coefficient, and $\beta$ is the wall reflection probability. In cylindrical coordinates, the lowest order solution is $\rho (r, t) = J_0 (\mu \frac{r}{r_0}) e^{-\gamma t}$ where $\gamma = \frac{\mu^2 D}{r_0^2}$, $\gamma_{\text{vol. quenching}}$ 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, $\gamma$ does not exhibit a $1/\text{pressure}$ dependence throughout the range $10 - 1000 \text{mm}^\text{Hg}$, since the boundary equation makes $\mu^2$ dependent on pressure and radius. At low pressure $\mu^2$ is linear in pressure.
and approaches a constant value. In the region between 10 - 700\,\mu\text{-}\text{Hg}, \mu^2 is a function of pressure and the diffusion decay rate does not have a 1/pressure dependence; only near one torr is \mu^2 a constant and behaves as 1/pressure.

The measured diffusion coefficient is $0.07 \pm 0.01 \, \text{cm}^2 \cdot \text{sec}^{-1}$ 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 $0.11 \, \text{cm}^2 \cdot \text{sec}^{-1}$ and $5.3 \times 10^{-15} \, \text{cm}^2$ respectively. The wall deexcitation probability, $(1 - \beta)$, is $0.22 \pm 0.08$ 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 $G_0^1$ state volume quenching rate; it is now measured as $335 \pm 5 \, \text{sec}^{-1} \, (\text{mm.} \cdot \text{Hg.})^{-1}$. 
WALL MATERIAL IS MYLAR.
WALL DE-EXITATION PROBABILITY IS 0.22.

\[ \gamma(p) - \gamma'(p) \]

\[ \gamma(p) \alpha \frac{1}{p} \]

TEST CELL RADIUS 0.75 cm

DECAY RATE \( \times 10^3 \) sec\(^{-1} \)

CO\(_2\) PRESSURE, microns-Hg

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APPENDIX B

CO₂ Relaxation Studies

Efforts have been made to examine the collisional decay modes of the $10^00$ vibrational level in the CO₂ 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 10.6 μm. 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 10.6 μm is proportional to the factor $(n_2 - n_1)$.

$$X(10.6) \propto (n_2 - n_1)$$

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 9.6 μm obtained from a separate Q-switch laser.

CO$_2$ Energy Levels

**FIGURE 1**
Fig. 2a  CO₂ Relaxation Data for 400 μ Hg
Fig. 2b  CO$_2$ Relaxation Data for 400 $\mu$Hg
Fig. 2c  CO$_2$ Relaxation Data for 400 $\mu$ 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 $\delta$ is the effective 9.6\(\mu\) pulse length, the non-equilibrium population densities are the following.

\[
\begin{align*}
\frac{n_1}{n_0}^{t_0+} &= \frac{n_3}{n_3}^{t_0+} = \frac{n_1^0 + n_3^0}{2} \\
\frac{n_2}{n_2}^{t_0+} &= n_2^0
\end{align*}
\]

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 10.6\(\mu\) before and after the 9.6\(\mu\) 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).
The diffusion coefficient for 001 state of CO₂ into CO₂ is measured and is shown to be twice larger than the self-diffusion coefficient of CO₂. 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 CO₂ amplifier saturation and manifestation of "Lamb dip" effect.
molecular relaxation
high power gas lasers
molecular laser amplifier