Dynamics of the CO$_2$ Lower Laser Levels as Measured with a Tunable Diode Laser*

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Abstract. A tunable diode laser operating in the 4.3 μm region is used to probe a conventional cw CO$_2$ laser discharge. Vibrational populations in the 10$^0$, 02$^0$, 02$^2$, and 01$^1$ levels of CO$_2$ are measured under lasing conditions, i.e., in the presence of intense 10.4 and 9.4 μm fields. The tunable diode laser is also used to monitor the energy transfer processes between the four levels after the passage of an intense 10.4 μm pulse. The detailed information provided by the tunable probe laser enables us to determine separately all the vibration-vibration (V-V) and vibration-translation (V-T) rate constants of importance in the relaxation of the lower laser levels in CO$_2$. The V-V rate constants are found to vary from a low value of $4.5 \times 10^4$ s$^{-1}$ Torr$^{-1}$ for the coupling of 01$^1$0 to 10$^0$ to a high value of $8.0 \times 10^5$ s$^{-1}$ Torr$^{-1}$ for the coupling of 01$^1$0 to 02$^2$0.

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The 10 μm CO$_2$ laser is one of the most powerful and efficient laser systems ever developed [1]. Consequently, the dynamics of the CO$_2$ laser have been studied in detail for many years. In general, there is now a good understanding of the behaviour of both pulsed and cw CO$_2$ lasers, and model predictions agree well with experiment. Nevertheless, there is still considerable uncertainty about the exact details of the relaxation mechanisms of the lower laser levels, 10$^0$ and 02$^0$ [2]. The development of 14 μm and 16 μm CO$_2$ lasers [3, 4] has led to renewed interest in the relaxation rates of these levels as such processes dominate the laser dynamics [4].

Much of the difficulty in determining the relaxation rates of the 10$^0$0 and 02$^0$0 levels arises from the fact that many different energy transfer processes are involved. First one must consider the vibration-vibration (V-V) processes involving the three levels 10$^0$, 02$^0$, and 02$^2$, i.e.,

\[
\text{CO}_2(10^0) + \text{M} \rightleftharpoons \text{CO}_2(02^0) + \text{M}, \quad (1)
\]
\[
\text{CO}_2(10^0) + \text{M} \rightleftharpoons \text{CO}_2(02^2) + \text{M}, \quad (2)
\]
\[
\text{CO}_2(02^0) + \text{M} \rightleftharpoons \text{CO}_2(02^2) + \text{M}, \quad (3)
\]

where M denotes any molecule in the gas mixture. All three levels are coupled to the 01$^1$0 level by

\[
\text{CO}_2(01^10) + \text{CO}_2(01^10) \rightleftharpoons \text{CO}_2(10^0) + \text{CO}_2(00^0) \quad (4)
\]
\[
\rightleftharpoons \text{CO}_2(02^0) + \text{CO}_2(00^0) \quad (5)
\]
\[
\rightleftharpoons \text{CO}_2(02^2) + \text{CO}_2(00^0). \quad (6)
\]

In addition to the V-V processes (1-6), all levels relax by the slower vibration-translation (V-T) processes represented by

\[
\text{CO}_2(01^10) + \text{M} \rightleftharpoons \text{CO}_2(00^0) + \text{M}. \quad (7)
\]

In view of the complexity of the lower laser level relaxation mechanism, one would ideally like to monitor the populations of all levels on an individual basis during the relaxation period. To date this has not been possible, and most experiments require many assumptions and approximations before relaxation rates can be extracted from laser pulse decays [5], fluorescence measurements [6], photo-acoustic measurements [7], and Raman scattering [8]. The most
direct experimental technique reported involved the use of saturating and probe lasers operating in the 10.4 and 9.4 μm bands [9, 10], but even these experiments required simultaneous account to be taken of the upper level population. It is not surprising that most experiments cannot differentiate between many of the processes (1–7), and often report relaxation rates for a given process which differ by an order of magnitude. The limitation of previous experimental techniques is perhaps best illustrated by pointing out that no measurements have ever been made on the 0220 level, despite its importance in the relaxation processes.

In this paper, we describe the use of a tunable diode laser to investigate the dynamics of the lower laser levels. The wide tunability of this probe laser enables us to monitor the populations in all levels of interest, including the 0220 and 0110 levels. We have measured the vibrational populations in typical cw CO2 discharges which are perturbed by intense 10.4 and 9.4 μm laser radiation. These measurements have revealed that the lower laser level relaxation rate is generally fast enough to maintain a Boltzmann distribution in the populations of the v1 and v2 modes. This is generally true, with the exception that at low gas pressures and high laser fields, the lower level directly pumped by the laser radiation has an anomalously large population. From the degree of perturbation of the pumped vibrational level, we can make a rough estimate of the effective relaxation rate.

A Q-switched CO2 laser is used to investigate the relaxation processes in more detail. The tunable diode laser monitors the population changes caused by the passage of an intense 10.4 μm pulse, and then determines the rate at which the various vibrational levels return to their equilibrium value. We have made measurements in both cw CO2 discharges and thermal gas mixtures over a wide range of pressures, and for various gas mixtures containing CO2, He, and N2. In particular, we have monitored the dynamics of the populations in the four levels 1020, 0220, 0210, and 0110, and developed a simple model, enabling us to relate the measured population changes to the rates of processes (1) to (7). This model is described in Sect. 4, and compared with experimental results in Sect. 5.

Sections 1 and 2 describe the experimental apparatus and the measurements made with cw laser radiation. The Q-switched measurements are described in Sect. 3, while Sect. 6 gives our conclusions.

This paper is a continuation of the work reported in two previous papers. The first paper described the use of a tunable diode laser to determine vibrational population distributions in a cw CO2 laser discharge [11], while the second paper investigated the dynamics of the upper laser level in the presence of intense 10μm laser radiation [12].

1. Experimental Apparatus

Figure 1 is a schematic diagram of the apparatus used for the cw measurements. A detailed description of the apparatus is given in previous publications [11, 12]; only a brief summary will be given here. The tunable diode laser emits in the 4.3 μm region and is used to probe transitions between CO2 levels (ijk) and (ijk+1) in a short discharge. Typical absorption bands probed by the tunable diode laser are indicated in Fig. 2. The short discharge tube has a 1 cm internal diameter and is placed inside a V-shaped laser cavity formed by two dichroic mirrors, M1 and M2, and a grating G. The majority of the incavity laser intensity is provided by a 1.0 m discharge. Thus, we are able to achieve a laser intensity of ~600 W/cm2 in the centre of the small discharge tube. This intensity produces a very large perturbation of the vibrational populations.

To investigate the relaxation processes of the lower laser levels the CO2 laser cavity is slightly modified. The gratings are replaced by a rotating mirror for Q-switching. The short discharge tube is now subject to a train of intense laser pulses with a peak power of ~5 kW/cm2, a pulsewidth at full-width-half-maximum of 160 ns, and a frequency spectrum distributed over the 10.4 μm P(16), P(18), P(20), and P(22) lines. To monitor the effect of these pulses, the chopper is removed from the probe beam, and the signal from the HgCdTe detector is fed directly into a storage oscilloscope with signal averaging capabilities (Tektronix model 468). The