Academic Research Staff

Prof. L. J. Chu Prof. C-I. Chang	Prof. H. A. Haus Prof. P. W. Hoff	Prof. J. A. Kong Prof. P. Penfield, Jr.
	Graduate Students	
E. L. Frohring T. Holcomb	D. L. Lyon A. H. M. Ross E. E. Stark, Jr.	W. A. Stiehl L. Tsang

A. SHORT PULSES PRODUCED BY CAVITY DUMPING OF A CO₂ TRANSVERSELY EXCITED ATMOSPHERIC (TEA) LASER

There is considerable interest in the production of single subnanosecond multimegawatt pulses of good mode quality from $\rm CO_2$ laser systems. These pulses could be used for the initiation of controlled fusion and for military applications. One approach involves the production of a short, high-power pulse, followed by a long high-pressure amplification stage. The initial pulse must have a fast rise time, high power, and good mode quality. The high power is needed so that nonlinear amplification can occur to combat gain narrowing. The fast rise time and mode quality are required for the intended applications, and the mode quality is also needed so that the cross-sectional beam size can be properly controlled in the amplifier stage and thereafter, As part of an ongoing program in this area, we report the production of single, short, high-power pulses from a $\rm CO_2$ TEA laser.

Pulses of 150-kW peak power, 20-30 ns long, were produced by a method of triggered cavity dumping (Fig. VI-1). Following a current pulse in the plasma tube, the buildup of the laser oscillation in the cavity is monitored, triggering the application of a quarter-wave voltage onto a GaAs crystal. This rotates the polarization of the laser field, causing the power to "dump" out of the cavity, off a germanium Brewster angle polarizer. The laser is 3.8 m long, contains a discharge tube 1 m long, and operates typically at 320 Torr total pressure. The dumped pulse passes through calibrated attenuators and is detected on a crystal of copper-doped germanium at 4°K.

Self-pulsing, caused by the oscillation of several longitudinal modes, occurs in the laser cavity, and this aids in the production of a short pulse for dumping (see Fig. VI-2.)

Switching external to the cavity can be an easier method of pulse production, but it acts only on that fraction of the internal power transmitted through an end mirror, and

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Fig. VI-1. System for production of short, intense pulses by cavity dumping of a $\rm CO_2$ laser.



1 A LONG

100 ns/div

20 ns/div





(b)

Fig. VI-2

- (a) Buildup of self-pulsed oscillation in the laser cavity, followed by dumping.(b) Dumped pulse, at 150 kW peak power.

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hence would produce lower power pulses from the same system.

Experimental studies of pulse amplification in low- and high-pressure regimes have been completed. Low-pressure studies have shown pulse shaping caused by the inertia of the macroscopic polarization as affected by nonlinear amplification. Pulse lengthening and nutation were observed in a pressure regime (above 5 Torr) where they have never previously been observed. High-pressure amplification studies have shown unexpected pulse shaping – higher gain in the trailing edge of the pulse than at the leading edge – because of fast equilibration of the very hot asymmetric stretch mode of vibration in CO_2 . These results give the first quantitative information about this equilibration process, and indicate that the asymmetric stretch mode relaxes significantly toward a Boltzmann equilibrium in less than 5 times the characteristic CO_2 - CO_2 intercollision time.

E. E. Stark, Jr., P. W. Hoff

B. PREDICTION OF GAIN IN CO₂ TEA LASERS

In previous reports^{1, 2} we have quantitatively described the dynamics of CO_2 TEA lasers. We introduce here a more concise formalism to express the flow of energy into and out of the active laser medium, which is composed of CO_2 , N_2 , and He gases. Comparisons are made between experimental determinations of small-signal optical gain and computer-generated calculations based on theory.

The earlier analysis¹ was based on laser rate equation formalism with selected vibrational eigenstates of the $\rm CO_2$ molecule used as basis states. In it we assumed initial vibrational temperatures. No explicit expressions were written to represent inelastic energy pumping through electron-molecule collisions. Nor did we consider interactions with N₂ molecules. In this report we organize the basis states differently and explicitly describe electron-neutral pumping and neutral-neutral interactions with N₂.

Rather than considering individual vibrational levels, we now treat entire modes of vibration. The vibrational mode of a perfectly harmonic vibrator has eigenstates equally spaced in energy according to the well-known prescription for its eigenvalues:

$$\mathscr{E}_{\xi} = \left(\xi + \frac{1}{2}\right) h\nu$$
 $\xi = 0, 1, 2, 3, ...,$

where $h\nu$ is the characteristic quantum of energy for the mode in question.

A simple harmonic mode in Boltzmann equilibrium at temperature T_v has an especially desirable property. The probability of occupation of any level and the energy associated with the mode are simple functions of the Boltzmann factor, $exp(-h\nu/T_v)$, where temperature is expressed in energy units. Consider a sample of diatomic molecules which vibrate in a perfectly harmonic manner. Assume a certain particle density, N.

The Boltzmann factor related to vibration of the molecules is written as $v = \exp(-h\nu/T_v)$. On the average, the number of molecules per unit volume occupying the kth excited vibrational level is N v^k(1-v). The energy per unit volume associated with vibration is N $h\nu v/(1-v)$, two familiar results of thermal physics.³

Each of the four simple vibrational modes of $\rm CO_2$ and the vibrational mode of $\rm N_2$ is considered to be perfectly harmonic and to be in Boltzmann equilibrium with separate vibrational temperatures. Three vibrational modes of $\rm CO_2$ (the symmetric stretching mode and two bending modes) are strongly coupled and are assumed to be characterized by a single temperature. We further assume that the characteristic quantum of symmetric vibration is exactly twice the bending quantum, and that the asymmetric quantum and the $\rm N_2$ vibrational quantum are exactly equal. Figure VI-3 shows the idealized mode structure in detail.



Fig. VI-3. Vibrational mode structure.

We assume the existence of several intermode interaction processes.⁴ Translation is coupled to the bending modes through the exchange of vibrational quanta for kinetic energy. This may occur during a collision of CO_2 with any other molecule. The probability per collision is denoted⁵ by $R_{k \rightarrow k \pm 1}$, under the assumption that the CO_2 molecule is originally in the kth level of bending vibration and finally in the k ± 1 level. The bending mode may exchange three quanta for a single asymmetric quantum, with probability $P_{\ell \rightarrow \ell \mp 3}^{k \rightarrow k \pm 1}$. The asymmetric mode exchanges single quanta with N_2 vibration with a

probability $Q_{\ell \to \ell \mp 1}^{k \to k \pm 1}$ for a CO_2 -N₂ collision, where the CO_2 molecule originally has ℓ asymmetric stretching quanta, and N₂ has k vibrational quanta. Each of these processes tends to bring the modes that are involved into thermal equilibrium at rates comparable to collision rates multiplied by a characteristic probability of interaction per collision.

At this time we are not considering mode interaction from stimulated transitions caused by intense coherent radiation fields. The probe beam used in the gain experiment is assumed to be very weak in intensity compared with the effective saturation parameter of the material.

The equations of motion for vibrational mode energies will now be developed. We characterize molecule-molecule vibrational energy exchange by probabilities of transition per collision. Using a one-dimensional exponential repulsion model developed by Schwartz et al., 6 we derive the dependence of the probabilities on initial and final vibrational quantum numbers.

We take as a specific example the asymmetric mode of CO_2 . The following equation expresses the time rate of change of asymmetric energy density from collisional interactions.

$$\frac{\mathrm{d}}{\mathrm{dt}} \mathscr{E}_{a} = \frac{2Z_{\mathrm{cm}}}{f_{\mathrm{c}}^{\mathrm{N}}} h \nu_{a} \left(\sum_{\mathrm{k}, \ell} P_{\ell \to \ell-3}^{\mathrm{k} \to \mathrm{k+1}} q_{\mathrm{k}} m_{\ell} - \sum_{\mathrm{n}, \ell} P_{\ell \to \ell+3}^{\mathrm{k} \to \mathrm{k-1}} q_{\mathrm{k}} m_{\ell} \right) \\ + \frac{Z_{\mathrm{cn}}}{f_{\mathrm{n}}^{\mathrm{N}}} h \nu_{a} \left(\sum_{\mathrm{k}, i} Q_{i \to i-1}^{\mathrm{k} \to \mathrm{k+1}} q_{\mathrm{k}} p_{i} - \sum_{\mathrm{k}, i} Q_{i \to i+1}^{\mathrm{k} \to \mathrm{k-1}} q_{\mathrm{k}} p_{i} \right),$$

$$(1)$$

where

 \mathscr{E}_{a} = energy density of CO₂ asymmetric stretching mode hv₂ = characteristic energy quantum of asymmetric mode

N = total gas particle density in the active medium

 f_{c} = mole fraction of CO₂ molecules

 $f_n = mole fraction of N_2 molecules$

 Z_{cm} = average collision rate of a CO₂ molecule with all collision partners

$$Z_{cn}$$
 = rate of CO_2 - N_2 collisions, proportional to density of N_2

 $P_{\ell \to \ell-3}^{k \to k+1} = \text{probability that a CO}_2$ molecule changes asymmetric quantum level from k to k+1 and bending level from ℓ to $\ell-3$

$$Q_{\ell \to \ell-1}^{k \to k+1} = \text{probability of transition between } k^{\text{th}} \text{ and } k^{\text{th}}+1 \text{ CO}_2 \text{ asymmetric levels, and} \ \ell^{\text{th}} \text{ and } \ell^{\text{th}}-1 \text{ N}_2 \text{ vibrational levels}$$

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$$\begin{split} \textbf{q}_k &= \text{density of CO}_2 \text{ molecules in } \textbf{k}^{\text{th}} \text{ asymmetric level} \\ \textbf{m}_{\textit{l}} &= \text{density of CO}_2 \text{molecules in } \textit{l}^{\text{th}} \text{ bending level} \\ \textbf{p}_{\textit{l}} &= \text{density of N}_2 \text{ particles in } \textit{l}^{\text{th}} \text{ vibrational level.} \end{split}$$

We define a, b, and n to be, respectively, the Boltzmann factors associated with CO_2 asymmetric, CO_2 bending, and N_2 vibrational modes. The following identifications can then be made:

$$\mathscr{C}_{a} = \frac{f_{c} N h \nu_{a} a}{1 - a}$$
$$q_{k} = f_{c} N a^{k} (1 - a)$$
$$m_{\ell} = f_{c} N b^{\ell} (1 - b)$$
$$p_{i} = f_{n} N n^{i} (1 - n).$$

The P's and Q's are now expressed in terms of vibrational level numbers associated with the transition.⁶ Detailed balancing at thermal equilibrium with translation (at temperature T) allows us to express probabilities of transition between levels x and y in terms of probabilities between y and x. Specifically,

$$P_{\ell+3 \to \ell}^{k \to k+1} = \frac{(k+1)(\ell+3)(\ell+2)(\ell+1)}{6} P_{3 \to 0}^{0 \to 1}$$

$$P_{3 \to 0}^{0 \to 1} = e^{-(h\nu_{a} - 3h\nu_{b})/T} P_{0 \to 3}^{1 \to 0} = e^{-\Delta/T} P_{0 \to 3}^{1 \to 0}$$
(2a)

and

$$Q_{\ell \to \ell-1}^{k \to k+1} = (k+1)(\ell) Q_{1 \to 0}^{0 \to 1}$$

$$Q_{1 \to 0}^{0 \to 1} = Q_{0 \to 1}^{1 \to 0}.$$
(2b)

Substituting the Boltzmann factor formalism and the relations (2a) and (2b), we arrive at the following equation:

$$\frac{d}{dt}a = -2Z_{cm}P_{0\to3}^{1\to0} \frac{(a-e^{-\Delta/T}b^{3})(1-a)}{(1-b)^{3}} - Z_{cn}Q_{0\to1}^{1\to0} \frac{(a-n)(1-a)}{(1-n)}$$

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Thus the equations of motion for vibrational energy reduce to equations of motion of Boltzmann factors.

It is also necessary to describe the transfer of energy to vibration effected during electrical discharge. We model the discharge between an individual resistor-pin electrode pair in the gain tube as a cylinder of radius, r, and height equal to the electrode spacing, z (see Fig. VI-4). Electric field, \mathscr{E}_{o} , kinetic energy distribution of the electrons, and all other discharge quantities are assumed spatially uniform and finite only inside of the discharge cylinder. For specific operating conditions of the TEA gain tube, time-dependent measurements of voltage and current have been made. Estimates of the cathode fall potential have been made using the theory of Ward.⁷ Subtraction of the cathode fall potential from measured voltage yields a net voltage between electrodes which can be converted in a straightforward manner to an E/N ratio by

 $E/N = \frac{Net Voltage Measured}{Electrode Separation \times Total Particle Density}$

The E/N ratio is a function of time which is found experimentally to be finite only for 1-2 μs after initiation of the discharge.



Fig. VI-4. Cylinder model of discharge geometry.

We then have, for specific operating conditions of the gain tube, time-dependent plots of E/N and total power dissipated during the discharge. Using calculations⁷ of electron energy distribution functions for our specific gas mixtures and range of E/N values, we are able to deduce the time-dependent power transfer to each of the

vibrational modes of interest.

Denoting the total power delivered to the asymmetric mode as $P_{a}(t)$, we find that

$$(f_c N)(z\pi r^2) \left(\frac{d\mathscr{E}_a}{dt}\right)_{due \ to} = P_a(t).$$

This yields the following equation of motion for a:

$$\frac{da}{dt} = \frac{(1-a)^2 P_a(t)}{f_c N \pi r^2 zh\nu_a}$$
$$-2Z_{cm} P_{0 \to 3}^{1 \to 0} \left(\frac{(a-e^{-\Delta/T} b^3)(1-a)}{(1-b)^3} \right)$$
$$-Z_{cn} Q_{0 \to 1}^{1 \to 0} \frac{(a-n)(1-a)}{(1-n)}.$$

It is relatively straightforward to write equations of motion for the other two Boltzmann factors and for T, the translational-rotational temperature, and we do not present those expressions here.

Rates appearing in the Boltzmann factor equations may be evaluated through careful comparison with published, experimentally derived, rate constants. 4

It is interesting to note the magnitudes of the various rates of interaction. At 350 Torr, total pressure with mixture ratio 12:2:1 :: $He:CO_2:N_2$ and translational temperature equal to 300 °K the following characteristic times apply:

 $\tau_{\rm BT}$ (Bending-Translation) ≈ 8 μs $\tau_{\rm AB}$ (Bending-Asymmetric) ≈ 20 μs $\tau_{\rm AN}$ (Asymmetric-N₂) ≈ 2 μs.

We have previously carried out small-signal gain experiments with a $\rm CO_2$ TEA pinresistor gain tube.² By relating measured optical gain to certain vibrational level populations we may test the present theory.

For input radiation of intensity I_{in} we find that

$$I_{out} = I_{in} e^{aL}$$

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Fig. VI-5. Gain for $He:CO_2:N_2$ mixture.



Fig. VI-6. Gain for He:CO_2 mixture.

for small-signal conditions, where L is the length of the tube in meters. Then α (per meter) may be expressed as

$$a = \frac{2r}{y} \frac{5.4 \times 10^{4}}{T} \frac{f_{c}}{(f_{c} + .76f_{n} + .7f_{h})} (1-b^{2})(1-b)^{2} (1-a)(a-b^{2}),$$

where r and y are indicated in Fig. VI-4.

In Figs. VI-5 and VI-6 comparisons between calculated and experimentally measured gain are plotted. Figure VI-5 applies to a mixture of He, CO_2 , and N_2 gases; Fig. VI-6 applies only to He and CO_2 . The discharge cylinder radius, r, was left as an adjustable parameter. Best fits were found for r = 1.5 mm with N_2 and 1.55 mm without N_2 . This value matches well with published determinations of the extent of discharge.⁸

Our next objective is to write a description of intense laser field interaction with the active medium. Electron power transfer for our experiment was calculated by W. L. Nighan⁹ of United Aircraft Research Laboratories, East Hartford, Connecticut. His generous assistance is much appreciated.

D. L. Lyon

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