

## Effect of organic additives on the upper laser level relaxation rate and the gain of a TE CO<sub>2</sub> laser

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**Abstract.** The vibrational relaxation rate of the upper laser level (00<sup>0</sup>1) in a TE CO<sub>2</sub> laser in the presence of xylene and tripropylamine is determined by means of experimental investigation of the time behaviour of the gain. It is shown that the presence of such additives in amounts which are optimal for TE CO<sub>2</sub> lasers has no influence on the vibrational relaxation rate of the 00<sup>0</sup>1 level. The increase in the electron excitation rate is found by experiment to be about 17% and 30% for xylene and tripropylamine, respectively. At the same time the duration of the excitation current pulses decreases considerably. The upper laser level population and the gain are calculated. The calculations show that the additives decrease the values of the 00<sup>0</sup>1 level population and the gain, which is in a good agreement with the experimental results.

### 1. Introduction

The effect of organic additives with a low ionisation potential on a cw CO<sub>2</sub> laser has been experimentally and theoretically investigated by Stefanov and Atanasov (1969) and Atanasov (1975). It is found that these additives increase the laser output power due to the change in the plasma parameters and depopulation of the lower laser level. The organic additives are also used to improve the homogeneity and stability of a TEA CO<sub>2</sub> laser discharge (Levine and Javan 1973, Boulanger *et al* 1973). This improvement causes an increase of the laser output parameters (Andrews *et al* 1978). It is found by Apolonov *et al* (1979) that the gain and the upper laser level population decrease as the tripropylamine pressure increases.

The purpose of this work is to determine the effect of tripropylamine and xylene on the upper laser level (00<sup>0</sup>1) lifetime by investigating the time behaviour of the gain in the presence of such additives. Also, the time dependence of the upper laser level population and the gain is obtained by using the theoretical model of Andrews *et al* (1975). In the calculation the influence of these organic additives on laser discharge parameters is taken into account.

### 2. Effect of organic additives on the upper laser level lifetime

For investigation of the effect of the additives on the upper laser level lifetime we used the method suggested by Reid *et al* (1972). This method consists in incorporating two (laser

and gain) sections in the laser cavity and studying the effect of the additional gain section on the time delay of laser pulses with respect to current pulses at different delays between the discharges in the two sections. For values of time considerably larger than the lifetime of the lower laser level and energy exchange time between  $\text{CO}_2$  and  $\text{N}_2$ , the effective lifetime,  $T_2$ , of the upper laser level is determined by the equation (Reid *et al* 1972)

$$\exp [-(t_0 + t_f'/2)T_2] = (K/g_{20})(t_f - t_f')/(t_f' - t_{th}') \quad (1)$$

where  $t_0$  is the time delay between current pulses in the two sections,  $t_f$  is the time delay between laser and current pulses in the laser section only,  $t_f'$  is the time delay between laser and current pulses when the two sections are fired,  $K$  is a constant,  $g_{20}$  is the peak small signal gain in the gain section, and  $t_{th}'$  is the time at which the increasing gain reaches the cavity losses.

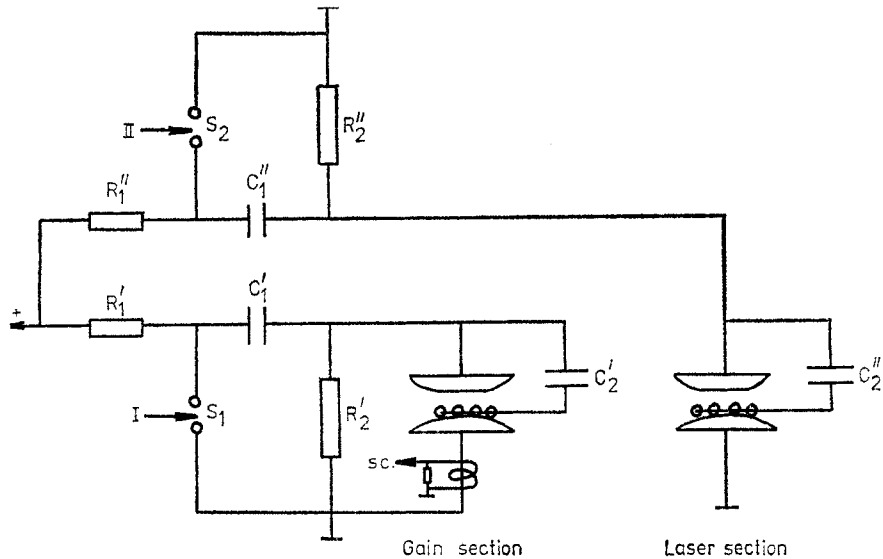


Figure 1. Electrical set-up;  $C_1' = 4$  nF,  $C_1'' = 10$  nF,  $C_2' = C_2'' = 2.8$  nF,  $U = 18$  kV–22 kV.

The electrical set-up is shown in figure 1. The two sections have identical Dumanchin construction (Dumanchin *et al* 1970). The main capacitors  $C_1'$  and  $C_1''$  are charged from 18 kV up to 22 kV from the same power supply and have values  $C_1' < C_1''$ . The pump energy densities of the gain and the laser sections at charging voltage 22 kV are  $74 \text{ J l}^{-1} \text{ atm}^{-1}$  and  $184 \text{ J l}^{-1} \text{ atm}^{-1}$ , respectively. The spark gaps  $S_1$  and  $S_2$  are fired with a time delay in the range 6–90  $\mu\text{s}$ . The capacitors  $C_2'$  and  $C_2''$  are used to supply the corona discharges. The laser cavity consists of two gold-coated mirrors with 2.4 m radius of curvature, at a distance of 119 cm. The laser output pulses are coupled out through the hole (3 mm diam) in the output mirror and are detected by a gold-doped Ge detector and a storage oscilloscope C8-2. The latter is switched on by means of a discharge current pulse in the gain section, which is fired first. Laser action from the gain section is eliminated through the small value of the capacitor  $C_1'$  and by introducing the calibrated losses in the cavity.

The results presented are for the gas mixture  $\text{CO}_2:\text{N}_2 = 1:1$  at total pressure of 100 Torr. The experiments are carried out in gas mixtures at these pressures, so that the

discharge has good stability and homogeneity both with and without additives. We consider this fact important for purer investigation of the influence of organic additives on the upper laser level lifetime, since Andrews *et al* (1978) report an increase of the laser output parameters in the presence of additives in the laser, but in the case without additives most probably the discharge does not have good homogeneity and stability and does not fill the whole volume between the electrodes.

We used various organic additives such as tripropylamine, xylene and ethyl alcohol. In our experiments the presence of these additives increased the peak current (i.e. the maximum value of electron density) and decreased the length of the current pulses and the peak gain.

The output pulses from the laser section with different delay with respect to the two discharges are shown on figure 2 (plate). The peak in the beginning of the picture corresponds to the discharge current in the gain section, which is not connected with the lasing. It can be seen that output pulses from the laser section, when the two sections are fired with different delays, have different amplitudes and delays,  $t_f'$ , with respect to the current pulses.

Equation (1) can be written as follows

$$\ln [(t_f - t_f') / (t_f' - t_{th}')] = \text{const} - [t_0 + (t_f' / 2)] / T_2 \quad (2)$$

from which  $T_2$  can be determined from the slope of the straight lines shown in figures 3(a, b, c). The slopes of the straight lines correspond to the values of  $T_2$ , which are in the range of accuracy ( $\pm 1 \mu\text{s}$ ) of the method. Consequently, as can be seen, the gain decay time, i.e. the upper laser level lifetime, is not changed in the presence of the additives. There is a good agreement between our data for the upper laser level relaxation rate without additives and those obtained by Reid *et al* (1972) and Tyte (1970).

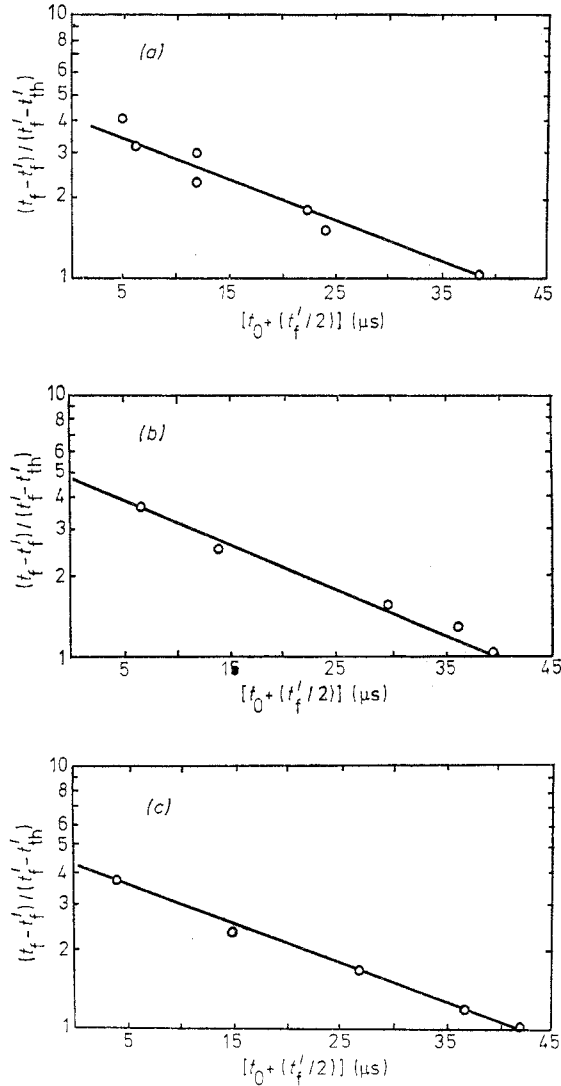
In short, it is seen from our results that the presence of the investigated additives in the above mentioned quantities does not affect the effective lifetime of the upper laser level. This conflicts with the results of Apolonov *et al* (1979).

Additional experiments show that for the pressures of the laser mixtures in which the discharge is stable and uniform without the presence of additives, the addition of the latter causes decrease of the gain. This fact is observed by Apolonov *et al* (1979), as pointed out before.

When tripropylamine and xylene are added the ratio of electric field intensity to total neutral particle number density decreases, i.e. the mean electron energy is reduced. At the same time the peak current increases by about 30% and 17%, respectively. The pulse duration decreases also compared with the case without additives. The current pulses are shown in figure 4 (plate) with tripropylamine (b), xylene (c) and without additives (a).

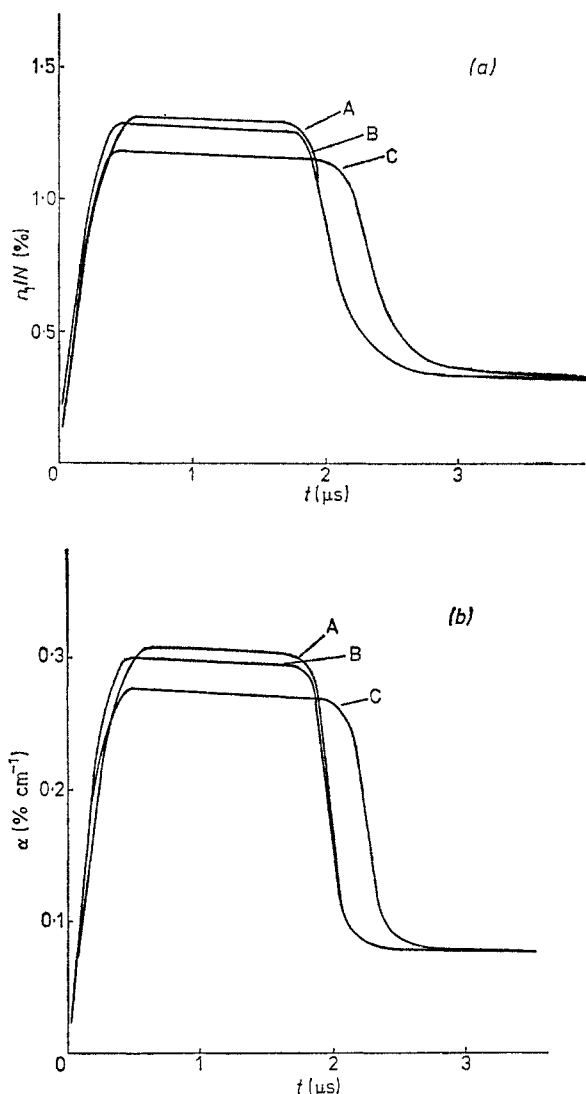
### 3. Calculation of the upper laser level population and the gain in the presence of additives

The calculations of the upper laser level population  $n_1$  and the gain coefficient  $\alpha$  as functions of time are carried out by means of the theoretical model given by Andrews *et al* (1975). On the basis of the experimentally obtained current (figures 4a, b, c) and voltage pulses the excitation pulse duration is determined and the maximum electron density and  $E/N$ —the ratio of electric field to neutral particle density—are calculated. The effective rate constants for the excitation of the upper laser level of CO<sub>2</sub> and the first vibrational



**Figure 3.** Decay of time dependent gain: (a) without additives; 50 Torr  $\text{CO}_2$ +50 Torr  $\text{N}_2$ ;  $p_\Sigma=100$  Torr.  $T_2=38 \mu\text{s}$ . (b) 50 Torr  $\text{CO}_2$ +50 Torr  $\text{N}_2$  with about 0.4 Torr tripropylamine;  $T_2=37 \mu\text{s}$ . (c) with about 1 Torr xylene;  $T_2=38.5 \mu\text{s}$ .

level of  $\text{N}_2$  are computed for  $\text{CO}_2:\text{N}_2=1:1$  mixture and for the obtained values of  $E/N$  according to Nighan (1970), Lowke *et al* (1973), and Engelhardt *et al* (1964). In the calculations, the influence of additives on the discharge parameters is taken into account. The presence of additives causes a small change in the form of the calculated electron energy distribution functions and the mean electron energy is between 1.7 eV in the presence of tripropylamine and 2.1 eV without additives. We found that the effective rate constants do not change significantly in this region of the electron energies, i.e. the change of the excitation rates of 00<sup>0</sup>1 level of  $\text{CO}_2$  and  $v=1$  level of  $\text{N}_2$  in presence of the additives is caused mainly by the change of electron density. We assume that the presence of additives at these pressures do not affect the upper laser level 00<sup>0</sup>1 lifetime, as our results



**Figure 5.** (a) The upper laser level population  $n_1$  and (b) the gain coefficient  $\alpha$  as a function of time. Curves: A,  $p_{\text{CO}_2}:p_{\text{N}_2}=1:1$ ,  $p_{\Sigma}=100$  Torr, without additives; B, with about 0.4 Torr tripropylamine; C, with about 1 Torr xylene.

showed. Moreover, we assume that tripropylamine and xylene do not affect the lower laser level since we have no knowledge of investigations asserting the opposite.

The results of the calculations are shown in figures 5(a), and (b). As can be seen, the population of the upper laser level and the gain decrease in the presence of the additives. The decrease is more pronounced for xylene than for tripropylamine. Decrease of the gain is also observed experimentally and there is a good agreement between theoretical and experimental results.

These results can be explained as follows. On the one hand, because of the increase of the peak current in the presence of additives, the maximum electron density is increased, i.e. the excitation rate of the upper laser level of CO<sub>2</sub> and the first vibrational

level of  $N_2$  are increased by 30% and 17% in the case of tripropylamine and xylene, respectively. On the other hand, the duration of the excitation pulses is decreased substantially, by about 30%. Therefore, the decrease of the excitation pulse duration in the presence of the additives has a stronger influence on the gain and the upper laser level population than the increase of the excitation rate of the upper laser level of  $CO_2$  and the first vibrational level of  $N_2$ .

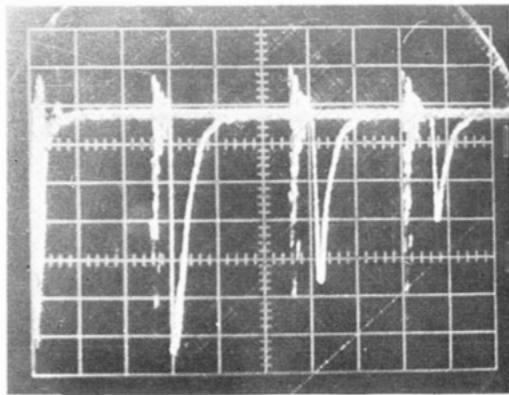
As can be seen from figures 5(a), and (b) population of the upper laser level and gain coefficient drop very rapidly at about  $2 \mu s$  because stimulated emission occurs. If the stimulated emission is not included in the calculation,  $n_1$  and  $\alpha$  should drop very slowly with a characteristic time equal to the observed time decay  $T_2$ .

#### 4. Conclusions

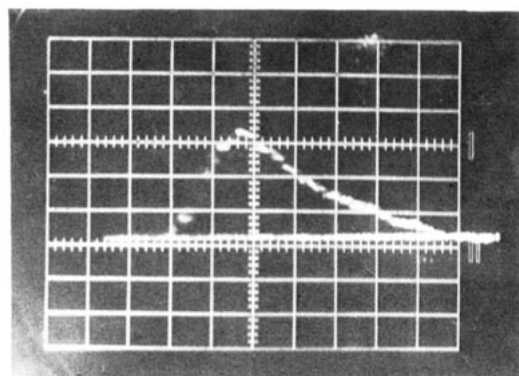
It is shown that the presence of tripropylamine and xylene in amounts which are optimal for TE  $CO_2$  lasers has no influence on the vibrational relaxation rate of the  $00^0_1$  level of  $CO_2$ . The increase of the electron excitation rate is found to be about 17% and 30% in the cases of xylene and tripropylamine, respectively. The calculations show that the additives decrease the  $00^0_1$  level population and the gain. This is in a good agreement with the experimental results.

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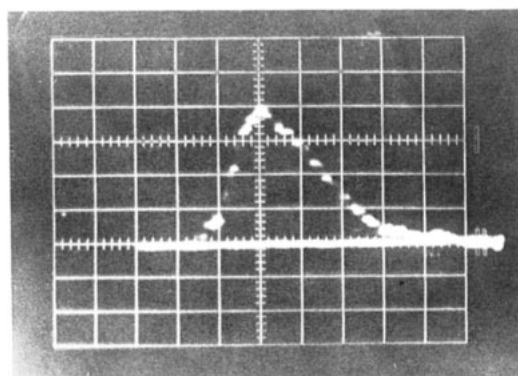
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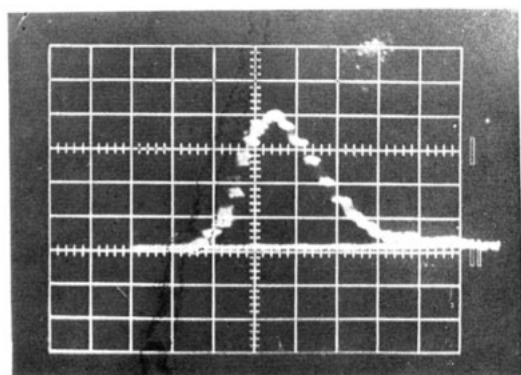
**Figure 2.** The output pulses from the laser section with different delay with respect to the two discharges. Time scale  $5 \mu\text{s cm}^{-1}$  reduced 60% in printing.  $p_{\text{CO}_2}:p_{\text{N}_2}=1:1$ ;  $p_{\Sigma}=100$  Torr.



(a)



(b)



(c)

**Figure 4.** Current pulses: (a)  $p_{\text{CO}_2} : p_{\text{N}_2} = 1 : 1$ ,  $p_{\Sigma} = 100$  Torr, without additives; (b) with about 0.4 Torr tripropylamine; (c) with about 1 Torr xylene. Time scale  $100 \text{ ns cm}^{-1}$ , reduced 60% in printing; current,  $80 \text{ A div}^{-1}$ ;  $U = 18 \text{ kV}$ .