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Comparison of models of CO_2 laser impedance fluctuations

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Abstract. There is a large opto-galvanic effect (OGE) in CO_2–N_2–He laser mixtures and this is exploited in laser frequency and power stabilisation systems. Two substantially different theories have been advanced to explain the effect. The two models are compared and it is concluded that the multi-step ionisation model is not adequate to describe the OGE in CO_2 lasers, but the temperature perturbation or discharge cooling model describes the phenomenon with considerable precision.

1. Introduction

The opto-galvanic effect (OGE) in CO_2 laser gas mixtures has required detailed examination because of its utilisation in laser frequency and power stabilisation. In the previous paper (Moffatt and Smith 1984, henceforth referred to as MS) we have presented a detailed analysis of the effect based on the discharge cooling mechanism established by Smith and Brooks (1979) and have found excellent agreement between the theory and experimental results. Alternatively Nowicki and Pienkowski (1982, henceforth referred to as NP) have advanced a substantially different theory of the OGE based on radiation field perturbation of the multi-step ionisation of the N_2 component usually present in CO_2–N_2–He laser gas mixtures. In this paper we compare these two models and conclude that the multi-step ionisation model as described by NP is not adequate to describe the OGE in CO_2 lasers.

Traditional models of the OGE have treated atomic systems where gas heating is small, excited levels (electronic) have energies which are a considerable fraction of the ionisation energy and the ionisation energy is much greater than the mean energy of the electrons in the excitation discharge. Hence two-step ionisation via an excited metastable level is usually dominant and the discharge voltage–current (V–i) characteristic exhibits a negative differential impedance slope. Theories such as that of Lawler (1980) associate the OGE with radiation field perturbation of the metastable level population and hence perturbation of the total ionisation rate. NP have constructed a model of the CO_2 molecular system based on such a perturbed ionisation sequence and the salient features of this model are outlined below in § 2.

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The OGE in molecular systems has become increasingly important not only for laser stabilisation (Moffatt and Smith (1981) and references therein) but also for tunable laser spectroscopy (see the recent review by Webster and Rettner (1983)). Discharges in gas mixtures including a molecular constituent generate considerable gas heating because of the relaxation of excited vibrational and rotational low-lying levels, and their negative *V–i* characteristics are usually attributed to decreased electron–neutral collisional energy losses as the number density decreases with increased gas temperature. The MS model of the OGE is based on the perturbation of the gas heating by the laser radiation field and this model is further compared with the NP mechanism in § 2 below.

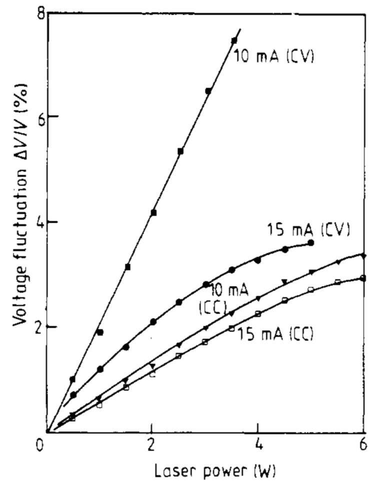

Figure 1. Variation of induced fractional voltage fluctuation $\Delta V/V$ with laser power switched for both constant current (CC) and constant voltage (CV) power supply operation at 10 mA and 15 mA in a mixture of 1 Torr Xe, 3 Torr CO_2, 6 Torr N_2, 18 Torr He.

It is convenient to here summarise some of the features of the experimental description of the OGE in CO_2 laser mixtures. Typically in a CO_2 laser if the internal radiation field is reduced without altering the discharge excitation (for instance by partially or completely blocking the radiation field or by misaligning a cavity mirror) the impedance of the discharge (as measured between the cathode and the anode) decreases. This is most commonly observed as a voltage decrease of up to $\approx$10%. Figure 1 shows the variation of the fractional voltage change $\Delta V/V$ with completely switched radiation field for a typical small laser (see MS for laser details). Results are presented for currents of 10 mA and 15 mA and with the power supply operated in both constant voltage (CV) and constant current (CC) modes of operation. The differences are due to feedback effects (both within the discharge and in the ballast circuit) and the gradient of the *V–i* characteristic, and have been examined in detail by Smith and Brooks (1979). Most of the feedback effects can be avoided, and the analysis simplified, if the discharge is operated in the constant current (CC) mode, and both MS and NP so operate.

Figure 2 shows the variation of several parameters with discharge current for a gas mixture of 3 Torr Xe, 3 Torr CO_2, 4 Torr N_2, 10 Torr He and the discharge power supply operated in the CC mode. The voltage (33 cm discharge length) decreases with current (curve A), the output power is a maximum in the 15 to 20 mA region (curve B) and the fluctuation $\Delta V/V$ is seen to have a maximum at a slightly lower current (10–15 mA)

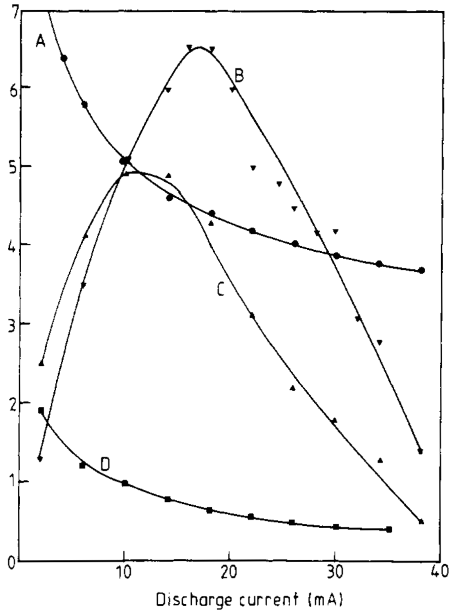

Figure 2. Variation with discharge current of (A) discharge voltage V (in kV), (B) laser output power P_{out} (in W), (C) voltage fluctuation $\Delta V/V$ (%) and (D) radiation field normalised voltage fluctuation $\Delta V/(VP_{out})$ (% W^{-1}) in 3 Torr Xe, 3 Torr CO_2, 4 Torr N_2, 10 Torr He.

(curve C). With constant current operation the voltage fluctuation $\Delta V/V$ is also the fractional impedance or resistance fluctuation $\Delta R/R$. $\Delta V/V$ (or $\Delta R/R$) can be normalised further to the change of laser power (P_{out}) to give $\Delta V/(VP_{out})$, which is a measure of the discharge change per unit change of radiation field. Strictly this should be $\Delta V/(VP_{lost})$ (where P_{lost} has been defined by MS, and is the power lost by the CO_2 system to the radiation field and is thus the sum of both P_{out} and the power lost in the cavity to diffraction, absorption and mirror losses etc), but this distinction is not important here. As seen in figure 2 (curve D), $\Delta V/(VP_{out})$ increases with a decrease of current and does not pass through a maximum, indicating that the OGE is most sensitive to radiation changes at the lowest operating currents (where the gradient of the V–i characteristic is greatest).

2. Summary of perturbation mechanisms

The MS theory has been detailed in the accompanying paper and is symbolically illustrated on the left of figure 3. It is assumed that an increase in the laser radiation field leads to a decrease in energy transferred to gas translational energy T_U by the relaxation process:

$$CO_2(001) + M \rightarrow CO_2\,(n, m, o) + M + T_U \qquad (1)$$

and hence a cooling of the gas and enhanced electron–molecule collision losses. In particular it is assumed that under typical laser device conditions there is an increase in the electron collision rate, but it is not necessary to assume any change in the energy exchange per collision. Thus when the laser field reduces the 001 population, the gas number density increases and the discharge impedance increases, as observed.

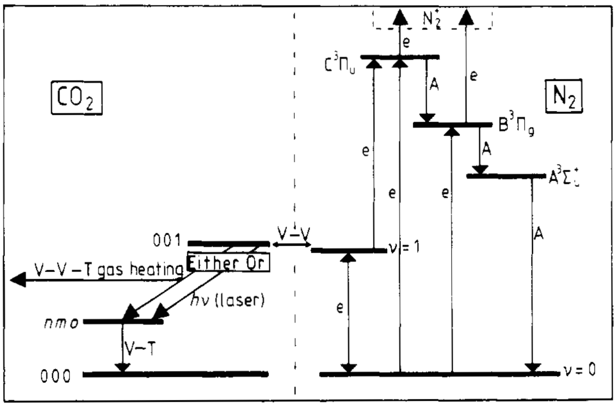

Figure 3. Selected portions of the CO_2 and N_2 energy level systems (not to scale), illustrating on the left the MS temperature perturbation model and on the right the NP nitrogen two-step ionisation model.

NP extend the traditional atomic multi-step ionisation model to the CO_2–N_2–He positive column discharge. They ignore any gas heating, but develop an extensive and complex rate equation model including all the most likely electron excitation, vibrational transfer, photon interaction and ionisation processes in CO_2, N_2 and CO. Both ionisation from the molecular ground states and from N_2 and CO electronic states are included, and charge loss is due to wall recombination. The electron density is determined from experimental *V–i* characteristics assuming literature values of the electron drift velocity, and likewise excitation and ionisation rates etc, are taken from the literature. It is not necessary to analyse this full and complicated model in detail, because, before it is applied, the authors simplify it considerably. First they maintain that the most important ionisation process is from the nitrogen $B^3\Pi_g$ state, and that all other ionisation processes are at least an order of magnitude less effective (NP p 1172). Further simplifying assumptions are made at equation (30). These are that the drift velocity, the reduced ambipolar diffusion coefficient and the ionisation rate coefficient of the $B^3\Pi_g$ state do not vary with electric field. In this way the NP model concludes that any voltage perturbation is directly and *pro rata* dependent on any $B^3\Pi_g$ state population perturbation, and such a perturbation only. In reality, drift velocity is nearly linearly proportional to the reduced electric field E/N (where N is the particle number density), and the ionisation rate coefficient varies very strongly with E/N (and in this model it is assumed constant). The salient features of the simplified model are shown on the right of figure 3: where e represents an electron impact process and A spontaneous relaxation. Note that any perturbation of the CO_2 upper laser level 001 is transferred to the nitrogen system by the fast V–V–T process:

$$CO_2(001) + N_2(\nu = 0) \leftrightarrow N_2(\nu = 1) + CO_2(000) \tag{2}$$

and then to the $B^3\Pi_g$ state *either* by electron excitation of the $N_2(\nu = 1)$ to the $C^3\Pi_u$ state and spontaneous relaxation to the $B^3\Pi_g$ state, *or* by electron excitation of the $N_2(\nu = 0)$ to the $B^3\Pi_g$. NP does not suggest which channel is dominant, but the following consideration is relevant. The $\nu = 0$ and $\nu = 1$ nitrogen ground-state populations will both change by the same total (*not* fractional) amount when the 001 population is varied, but with opposite signs. With laser action reducing the 001 population the N_2 $\nu = 1$ and $C^3\Pi_u$ populations will also decrease and hence the $C^3\Pi_u$ channel will tend to a decrease of the

$B^3\Pi_g$ state population, reduced ionisation and increased impedance (as observed). Whereas the accompanying increase in the N_2 $\nu = 0$ population should lead to enhanced direct excitation of the $B^3\Pi_g$ state and reduced impedance, contrary to what is observed experimentally. So one must assume the first channel is dominant.

In the following sections we first consider in detail various stages in the NP mechanism model and then we compare the model with experimental results.

3. Detailed consideration of the proposed ionisation mechanism channel

3.1. *Frequency effects*

It would normally be appropriate at this stage to quantitatively examine the NP model. Unfortunately even the simplified model is complicated and the authors have only given a brief outline of the utilisation of their model numerically. So for consideration one has to rely on assessment of certain prominent features or crucial steps, and examination of the few results presented.

It has been experimentally established (Smith and Moffatt 1979, Moffatt and Smith 1981, Schiffner 1971) that in a CO_2–N_2–He mixture the amplitude of the OGE falls off with increase of frequency of the cavity field modulation, approaching zero in the 1–2 kHz region. But the OGE returns at higher frequencies with opposite polarity, reaching a new maximum at ≈20 kHz and finally disappearing at ≈100 kHz. The exact frequencies depend on the gas mixture and total pressure. Moffatt and Smith (1981) have explained these features in terms of the rate of V–V–T relaxation of the upper laser level (equation (1)) and the rate of relaxation of the lower laser level system. Leaving aside this interpretation of the frequency effects there are consequences of the experimental results, such as:

(i) Any model must allow for the changing amplitude response with frequency. Thus NP obtain their experimental results with a cavity chopping frequency of 300 Hz, when for their gas mixture and pressure the amplitude response will be ≈50% of the response with slow modulation. This is of importance for numerically matching results (see § 3.4).

(ii) It is difficult to see how the NP model can account for the sign reversal with frequency increase above 1–2 kHz.

(iii) The vibrational transfer reaction (2) has a rate dependent on the gas composition and total pressure (Rosser *et al* 1969, Guegin *et al* 1975). For the appropriate gas mixture and estimated gas temperature the transfer should be considerably reduced at 20 kHz and approach zero at a frequency much less than the observed cut-off of ≈100 kHz. The process does not appear fast enough for an essential step in the mechanism channel; it would have to be at least 3 to 5 times faster.

3.2. *Lifetime of the $B^3\Pi_g$ state*

Central to the NP interpretation is the dominant ionisation from the $B^3\Pi_g$ electronic state. NP have based this section of their work on the theoretical work of Aleksandrov *et al* (1978). Aleksandrov *et al* predict rates for a pure nitrogen plasma and obtain good agreement with experimentally observed total ionisation coefficients etc, but make no comparison with results for a glow discharge. Note:

(i) Application of these results to a CO_2–N_2–He mixture is likely to lead to error by at least an order of magnitude (Sakai *et al* 1979).

(ii) There are two channels within the model by which the $B^3\Pi_g$ state can lose energy, by ionisation and by spontaneous decay. The $B^3\Pi_g$ state decays with a rate of $8.3 \times 10^5\,s^{-1}$. (Carleton and Oldenberg (1962). Other authors have slightly different values, but all are within the range 10^5–$10^7\,s^{-1}$.) So if ionisation from this state is to be significant, its rate must be at least of comparable magnitude. From table 1 of Aleksandrov *et al* (1978) the ionisation rate constant can be obtained for any particular E/N value. The E/N for a discharge in pure nitrogen or in the laser mixture will be within the range $2 \times 10^{16}\,V\,cm^2$ to $5 \times 10^{16}\,V\,cm^2$, and the corresponding rate constants will then be $4.7 \times 10^{-16}\,cm^3\,s^{-1}$ and $2.8 \times 10^{-10}\,cm^3\,s^{-1}$. Assuming an electron density of $\sim 10^{10}\,cm^{-3}$ the ionisation rate will be between 4.7×10^{-6} and $2.8\,s^{-1}$. Even allowing for an increase of rate in a CO_2–N_2–He mixture rather than in pure nitrogen ((i) above) it appears that the spontaneous relaxation will be totally dominant and the $B^3\Pi_g$ state is effectively not metastable and is not involved in a two-step ionisation process.

(iii) The theoretical analysis of Aleksandrov *et al* (1978) then does not support two-step ionisation via the $B^3\Pi_g$ state as being dominant. But it is interesting to note that Wiegand *et al* (1972) in their analysis of the nitrogen discharge have concluded that two-step ionisation via the $A^3\Sigma_u^+$ is important. This is much more likely because the $A^3\Sigma_u^+$ state is long lived with a lifetime of about 2 s (Carleton and Oldenberg 1962) and so at some possible E/N values ionisation from it may take place at a rate comparable with its relaxation.

3.3. The discharge V–i *characteristic*

The potential difference between the anode and the cathode is determined both by the charge creation and loss processes and by the (other) energy loss processes. If the ionisation process is significantly changed there should be a modification of the V–i characteristic. In figure 4 are shown the V–i characteristics for a typical laser mixture of 3 Torr CO_2, 4 Torr N_2 and 17 Torr He, and the identical mixture with the nitrogen replaced by 4 Torr of carbon monoxide. The mixture with the CO requires a slightly lower potential difference to maintain a particular current. This is completely at variance with the NP theory of the two-step ionisation within the nitrogen component being dominant for then there should be a considerably lower impedance discharge with the

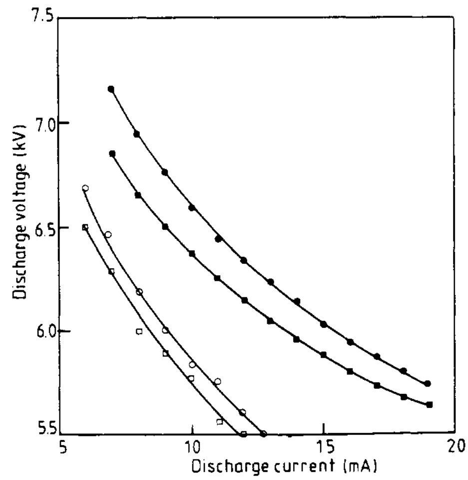

Figure 4. Variation of discharge voltage with discharge current (33 cm discharge length) in 3 Torr CO_2, 17 Torr He with added 4 Torr N_2 (●), 4 Torr CO (■), 4 Torr N_2 plus 1 Torr Xe (○) and 4 Torr CO plus 1 Torr Xe (□).

nitrogen present. Indeed such an impedance decrease is easily observed if a low ionisation-potential additive such as xenon is added to the gas mixture (figure 4).

3.4. Perturbation predictions

The only current perturbation results which NP present are in their figures 2 and 3. Consider figure 3 first. This shows the shape of the laser power output curve is the same as the shape of the calculated impedance fluctuation $\Delta R/R$ (and our $\Delta V/V$—see curve C of figure 2 above) as the cavity losses are changed by changing the laser output coupling. As NP observe, this establishes (in the first approximation) that the impedance fluctuation depends linearly upon the photon density. Nothing further is established about the OGE mechanism.

In figure 2 NP show how the laser output power (their P_L, our P_{out}) varies with discharge current, and also how both the theoretical and experimental impedance perturbations vary with current. Note that the impedance perturbation results are presented as a correlation coefficient $\eta = (\Delta R/R)(\Delta n_f/n_f)^{-1}$ where n_f is the cavity photon density. But since the results are obtained by turning on and off the radiation field by an intracavity chopper (at 300 Hz), presumably $\Delta n_f = n_f$ and η does not involve any normalisation to the radiation field. The experimental fluctuation maximises in the 10–12 mA region, in a generally similar manner to curve C of figure 2 of this paper. The theoretical curve fits the experimental points very well, both in the shape and amplitude. But it is not clear from the text if the theoretical curve has been normalised to the experimental points or not. It is probable that it has been, for §§ 3.1 (i) and 3.2 (i) above in particular would suggest that any exact amplitude fit would be coincidental.

Consider the shape of the impedance fluctuation. In § 1 above we pointed out that the most physically significant parameter is not the fluctuation $\Delta V/V$ or $\Delta R/R$, but the fluctuation normalised to the radiation field intensity $\Delta V/(VP_{out})$. In NP this would be $(\Delta R/R)(P_L)^{-1}$ and if the impedance fluctuation results of figure 2 of NP are replotted as the variation of $(\Delta R/R)(P_L)^{-1}$ with current a curve very similar to that of our figure 2, curve D is obtained. There is no longer a maximum, but the normalised fluctuation is greatest at the lowest current.

To summarise, the NP results as presented in their figures 2 and 3 establish the same

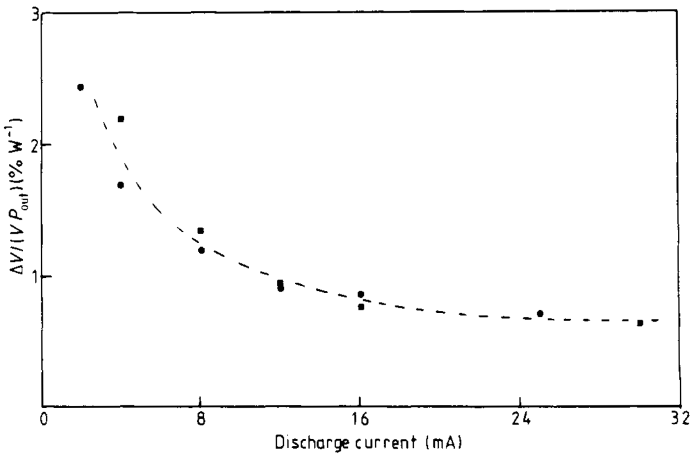

Figure 5. Variation of field normalised voltage fluctuation $\Delta V/(VP_{out})$ with discharge current in 1 Torr Xe, 3 Torr CO_2, 4 Torr CO, 17 Torr He (●), and in 1 Torr Xe, 3 Torr CO, 4 Torr N_2, 16 Torr He (■).

general behaviour as observed by MS and others previously, the effect is dependent on the radiation field intensity and is largest at low currents. That the effect should be largest at low current is to be expected on an ionisation model because any ionisation change will then have the most significant effect on the discharge. Likewise on the MS model the OGE will be greatest at low current because the discharge power (heating) is then most sensitive to a change in power (heating).

3.5. The amplitude of the OGE

In a previous paper (Smith and Brooks 1979) we have shown that there is a large OGE in a pure CO_2 discharge as well as in CO_2–N_2–He mixtures, but of opposite polarity because the CO_2 is usually in a state of absorption rather than emission. Figure 5 shows results for the variation of the normalised effect $\Delta V/(VP_{out})$ with current for a laser mixture including nitrogen (squares) and a similar mixture excluding nitrogen but including carbon monoxide (circles). Clearly the presence or absence of nitrogen has very little effect on the amplitude of the OGE.

4. Conclusions

The opto-galvanic effect in typical CO_2–N_2–He–(Xe) gas mixtures is not dependent on the presence of the nitrogen component (§ 3.5), and the N_2 does not appear to play any significant role in maintaining the electrical discharge (§ 3.3). The proposed nitrogen two-step ionisation mechanism (Nowicki and Pienkowiski 1982) for opto-galvanic perturbation is not sufficient to explain the OGE in the CO_2 laser (§§ 3.1, 3.3). The Nowicki and Pienkowski model has several basic shortcomings (§ 2) and the experimental evidence produced to substantiate it, while in qualitative agreement with other workers, is not specific to the particular model (§ 3.4), whereas the temperature perturbation (MS) model can predict all the reported results of this phenomenon with considerable precision.

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