

Physics of CO₂ Electric Discharge Lasers

R. H. BULLIS,* W. L. NIGHAN,† M. C. FOWLER,‡ AND W. J. WIEGAND§

United Aircraft Research Laboratories, East Hartford, Conn.

Detailed analytical and experimental investigations of the major factors which influence the performance of CO₂ electric discharge lasers have been conducted. Rates of electronic excitation of key laser levels have been computed from analytically determined electron energy distributions which are non-Maxwellian in CO₂ laser discharges. Combination of these data with a model of molecular kinetic processes has permitted quantitative calculation of important laser properties such as gain and saturation intensity as a function of various discharge parameters. Further, analysis of discharge plasma properties has made possible comparison of calculated results with experimental data yielding both qualitative and quantitative agreement. These results reveal the dependence of laser properties on discharge characteristics, thereby illuminating the central features of CO₂ laser operation.

Nomenclature

u = electron energy in eV

$f(u)$ = electron energy distribution function normalized so that

$$\int_0^{\infty} u^{1/2} f(u) du = 1$$

$\bar{u}_r$ = reduced average electron energy defined as

$$\frac{2}{3} \int_0^{\infty} u^{3/2} f(u) du$$

E/N = ratio of electric field intensity (E) to total neutral particle number density (N)

T = gas translational temperature

T_v = vibrational temperature characteristic of N₂

T_1 = vibrational temperature characteristic of coupled CO₂ bending and symmetric stretch modes (lower laser level system)

T_2 = vibrational temperature characteristic of CO₂ asymmetric stretch mode (upper laser level system)

n_e = electron number density

N_{001} = number density of CO₂ upper laser level

N_{100} = number density of CO₂ lower laser level

I_s = saturation intensity for 10.6 μ radiation

E/P = ratio of discharge electric field intensity to total gas pressure (P)

IP = product of discharge current (I) and total pressure

PR = product of total pressure and radius (R) of a wall dominated, cylindrical discharge column

σ = cross section for stimulated emission of 10.6 μ CO₂ laser radiation

α_o = small signal gain coefficient

metric stretch-N₂ vibrational systems, combined with selective relaxation of the coupled CO₂ symmetric stretch-bending vibrational systems.¹ The usefulness of the CO₂ discharge as a laser medium is determined in large measure by the magnitude of the resulting asymmetric stretch-symmetric stretch vibrational population inversion and the ability of the system to sustain the population inversion as the strength of the generated 10.6 μ optical field increases. Clearly, an understanding of the details of the physical phenomena influencing CO₂ laser performance requires a study of the factors influencing electron, molecular, and plasma kinetic processes in laser discharges.

Electron kinetic processes in CO₂ laser discharges are particularly difficult to analyze because of the numerous vibrational and electronic excitation processes of importance in mixtures of molecular gases. Evaluation of electron transport coefficients and excitation rates is further complicated by the fact that electron energy distributions are non-Maxwellian under conditions typical of CO₂ laser operation. Recently, Nighan²⁻⁴ reported results of an analytic study of electron kinetic processes in CO₂ laser mixtures which permit quantitative evaluation of important electron-molecule excitation rates for the conditions of interest in CO₂ laser discharges. Section II of the present paper summarizes the essential details of this study. Using these calculated electron-molecule rate constants along with those for molecule-molecule energy relaxation, Fowler⁵ has developed a model of CO₂ molecular kinetic processes. This analysis permits quantitative evaluation of the influence of plasma properties on gain and saturation intensity in electric discharge lasers, and is reviewed in Sec. III.

The plasma physics of the electric discharge plays an important role in CO₂ laser performance. The conclusions of Secs. II and III reveal that average electron energy, electron density, and gas temperature are of prime importance to laser operation. Therefore, the factors influencing these discharge properties must be clearly identified. Section IV of this paper describes the details of an analysis of positive column properties in conventional, wall-dominated CO₂ laser discharges. From this study, the importance of ionic species and effective ionization rates are determined and the plasma electron density and gas temperature predicted for a variety of discharge conditions.

In Sec. V the results of Secs. II-IV are combined to provide an internally self-consistent prediction of small signal gain in a CO₂-N₂-He laser discharge. These results are compared with experimental measurements of small signal gain obtained for similar conditions. The comparison reveals excellent qualitative and quantitative agreement, providing evidence that the important physical processes occurring in CO₂ lasers have been modeled correctly. The analytical results described herein are therefore useful in situations where extensive experimental observation is not practical and suggest techniques for over-all improvement in CO₂ laser performance. The implications of these studies regarding high power CO₂ lasers are discussed in Sec. VI.

I. Introduction

THE efficient conversion of electrical energy to optical energy typical of electrically excited CO₂ lasers has stimulated considerable interest in the physics of CO₂ laser discharges. Basically, a vibrational population inversion is produced in electrically excited CO₂ laser mixtures, usually composed of CO₂, N₂, and He, by selective electron excitation of the coupled CO₂ asym-

Presented as Paper 71-64 at the AIAA 9th Aerospace Sciences Meeting, New York, January 25-27, 1971; submitted March 10, 1971; revision received December 17, 1971. Portions of this work were supported by the Office of Naval Research.

Index categories: Atomic, Molecular and Plasma Properties; Lasers.

* Principal Scientist, Plasmadynamics. Member AIAA.

† Senior Research Scientist, Plasmadynamics.

‡ Experimental Physicist, Gas Laser Physics.

§ Research Scientist, Plasmadynamics.

II. Electron Kinetics

The high efficiency of CO_2 - N_2 -He gas discharge lasers reflects the very effective energy exchange between the plasma electrons and the relevant vibrational levels of CO_2 and N_2 . The cross sections for vibrational excitation of CO_2 and N_2 by electron impact are indeed found to be relatively large.^{6,7} As might be expected, the reported data indicate that the number of molecular energy levels participating in electron collisional processes is likely to be very large in CO_2 laser mixtures. In addition, low pressure electric discharges in molecular gases are known to exhibit non-Maxwellian electron energy distributions.²⁻⁴ For this reason it is clear that estimates of electron-molecule excitation rates based on assumed values of "electron temperature" are likely to be erroneous. Therefore, considerable analysis is required to develop an understanding of electron energy transfer processes in CO_2 lasers. Accordingly, electron energy distribution functions were obtained by numerically solving the Boltzmann equation for CO_2 - N_2 -He mixtures.²⁻⁴ These distributions were then used to compute electron energy transfer rates for the various vibrational and electronic levels of CO_2 and N_2 .

Electron Energy Distributions

In order to determine the electron energy distribution function, it is necessary to solve the Boltzmann equation taking into account the effect of electron-neutral, electron-ion, and electron-electron collisions. Solution of this equation requires knowledge of all the important electron scattering cross sections over the energy range of importance (approximately 0–20 eV for CO_2 lasers). A comprehensive discussion of the cross sections used in the analysis of electron energy distributions is reported by Nighan.⁴

Using available electron cross section data,⁶⁻⁸ the electron kinetic equation was solved numerically with E/N , the fractional species concentrations, and the fractional ionization taken as variable parameters. Calculations were made over a range of these parameters typical of CO_2 gas discharge lasers. Shown in Fig. 1 are electron energy distribution functions for various E/N

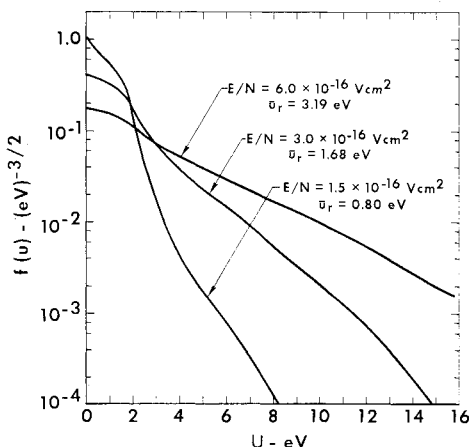


Fig. 1 Computed electron energy distributions for a CO_2 - N_2 -He mixture having the number density proportions 0.1–0.1–0.8, respectively.

values calculated for a CO_2 - N_2 -He mixture in the proportions 0.1–0.1–0.8 by number density. For values of fractional ionization (n_e/N) in the 10^{-8} – 10^{-6} range typical of laser operation, Coulomb collisions were found to have no significant effect on the distribution function. Furthermore, when the effect of electron super-elastic collisions with vibrationally excited CO_2 and N_2 was taken into account, the distribution function was not affected substantially for excited state populations corresponding to vibrational temperature levels common to CO_2 laser discharges.

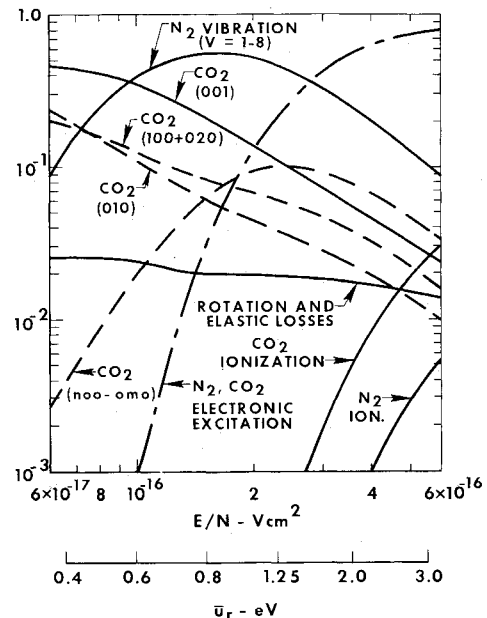


Fig. 2 Fractional electron power transfer for the conditions of Fig. 1. For clarity of presentation, the electron power input to several levels has been summed as described in Refs. 2–4.

Electron Fractional Power Transfer

With a knowledge of the electron energy distribution function, it is possible to perform an energy balance by integrating the electron kinetic equation over all electron energies.⁴ This procedure leads to an electron energy conservation equation which equates the rate of energy input to the electrons from the electric field to the rate of electron-molecule energy transfer, from which the relative contribution of the various electron energy loss processes is readily obtained. The resulting fractional electron power transfer is closely related to laser efficiency. Figure 2 shows the fractional power transferred from the electrons to CO_2 , N_2 , and He as a function of E/N and $\bar{u}_r$, the reduced average energy.⁴ For clarity of presentation, the power input to several levels has been added as indicated. For this mixture and an E/N value of approximately $8 \times 10^{-17} \text{ V cm}^2$, 40% of the electron energy is transferred directly into the CO_2 upper laser level (001) and effectively all the energy goes into vibrational excitation of either CO_2 or N_2 . The relatively large low energy cross section for electron excitation of the CO_2 (001) level is responsible for the dominance of upper laser level excitation for values of $\bar{u}_r$ near 0.5 eV. On the other hand, for an E/N value of about $6 \times 10^{-16} \text{ V cm}^2$, approximately 80% of the electron energy is transferred to electronic excitation of CO_2 and N_2 . These results indicate that the $\bar{u}_r$ range 0.5–2.0 eV is a transition region for CO_2 laser mixtures, in which the dominant electron energy transfer process changes from vibrational excitation to electronic excitation. Clearly, electron energy loss associated with elastic collisions and with those resulting in rotational excitation and/or ionization is insignificant (Fig. 2). Nevertheless, in order to sustain the discharge in the presence of charged particle losses, ionizing collisions are required. For most CO_2 laser conditions the required ionization rate is provided when $\bar{u}_r$ is approximately 1.0 eV. For this situation, the data of Fig. 2 indicate that approximately 70% of the electron energy is transferred through the CO_2 (001)- N_2 ($v = n$) system. After consideration of the 41% quantum efficiency of the CO_2 (001)–(100) laser transition, this suggests that for $\bar{u}_r \sim 1.0$ eV nearly 30% of the electrical energy is potentially available for conversion to 10.6μ optical radiation.

III. Molecular Kinetics

Knowledge of the vibrational energy transfer rates for electron-molecule and molecule-molecule collisions permits calculation

of the steady state CO₂ vibrational level populations for conditions typical of CO₂ electric discharge lasers. Electron-molecule rate coefficients can be computed for a variety of circumstances by employing the electron distribution functions as described in Sec. II along with the appropriate cross sections. Reliable molecule-molecule rate coefficients have been reported in the literature.⁹ These data can be used in conjunction with a molecular kinetics model for gas mixtures and plasma conditions typical of CO₂ electric discharge lasers. Details of the analytic modeling procedure and important rate processes have been described previously⁵ and are summarized in the following paragraphs.

Kinetic Model

The kinetic models used for CO₂ and N₂ are very similar to those proposed by Gordietz et al.¹⁰ and by Moore et al.¹¹ The usual notation for the vibrational state of CO₂ is utilized where l , m , and n are the vibrational quantum numbers for the symmetric stretch, bending, and asymmetric stretch modes, respectively. Each mode of vibration is considered to be a simple harmonic oscillator. Consequently, intramode vibrational energy transfer is resonant in character, occurring at a rate close to the value of the gas kinetic frequency, and is therefore much faster than all the other energy transfer processes considered.¹² Hence, the energy distribution within a mode is Maxwellian and can be characterized by a temperature. In addition, the symmetric stretch and bending modes of CO₂ are coupled both collisionally and by the Fermi resonance which exists between them,¹³ so that the energy distribution in these modes can be characterized by a single vibrational temperature. Therefore, in an electric discharge containing CO₂ and N₂, there are three vibrational temperatures of interest: T_2 characterizing the CO₂ asymmetric stretch mode, T_1 the coupled CO₂ symmetric stretch and bending modes, and T_v the N₂ vibrational system.

Collision Processes

There are seven basic collisional and radiative processes influencing the CO₂ and N₂ vibrational level populations for conditions typical of electrically excited CO₂ lasers. These are illustrated schematically in Fig. 3 and may be summarized as follows: 1) electron vibrational excitation and de-excitation of the CO₂ asymmetric stretch vibration⁴; 2) vibrational energy exchange between N₂ and the CO₂ asymmetric stretch mode⁹; 3) collisional de-excitation (and excitation) by atoms and molecules of the CO₂ asymmetric stretch vibration⁹; 4) electron

vibrational excitation and de-excitation of N₂ (Ref. 4); 5) excitation and de-excitation by both electrons and heavy particles of the coupled CO₂ bending and symmetric stretch modes^{4,9}; 6) CO₂ spontaneous emission of 4.3 μ radiation; and 7) CO₂ stimulated emission and absorption of 10.6 μ radiation. Using the harmonic oscillator approximation,¹⁴ the rate constants for these processes were averaged over all the appropriate vibrational levels, giving rate constants dependent on the characteristic vibrational temperatures. The value of the cross section for stimulated emission of radiation was found to be very important in calculating the value of the rate constant for process 7. The dependence of the stimulated emission cross section on gas temperature and pressure was obtained using the methods described previously⁵ and is shown in Fig. 4 for a typical CO₂ laser mixture.

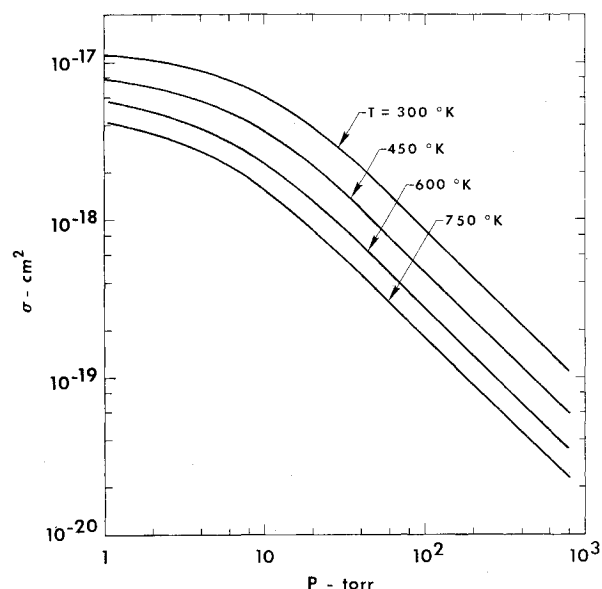


Fig. 4 Cross section for stimulated emission for the $P(20)$ line of the 001-100 CO₂ laser transition computed for a CO₂-N₂-He 0.1-0.1-0.8 laser mixture using the methods described in Ref. 5. Since the cross section is not particularly sensitive to mixture variations, these data are representative of many CO₂ laser discharges.

The asymmetric stretch-symmetric stretch population inversion which gives rise to 10.6 μ radiation emission was calculated directly from the solution to the equations representing steady state conditions. The gas temperature and electron density were taken as independent parameters in the calculation. Although these plasma properties cannot be independently controlled under ordinary discharge conditions, in the present work they were treated as independent variables in order to evaluate the parametric dependence of laser performance on a given property.

Laser Gain and Saturation Intensity

The gain in a CO₂ laser is equal to the product of the cross section for stimulated emission and the CO₂ (001)-(100) population inversion, and represents the fractional increase in the 10.6 μ radiation intensity per unit path of laser medium traversed by the radiation. The value of gain in the limit of zero radiation field is the small signal gain, α_0 , whereas the value of the radiation intensity at which the gain has been reduced to half its small signal or unperturbed value is defined as the saturation intensity. The product $\alpha_0 I_s$ is representative of the maximum available optical power density of the medium and is therefore useful in evaluating the influence of various discharge properties on laser performance.

Values of small signal gain and saturation intensity have been calculated for a range of electron densities, gas temperatures, and values of radiation field intensity. These calculations were performed using representative values of E/N and for gas pressures

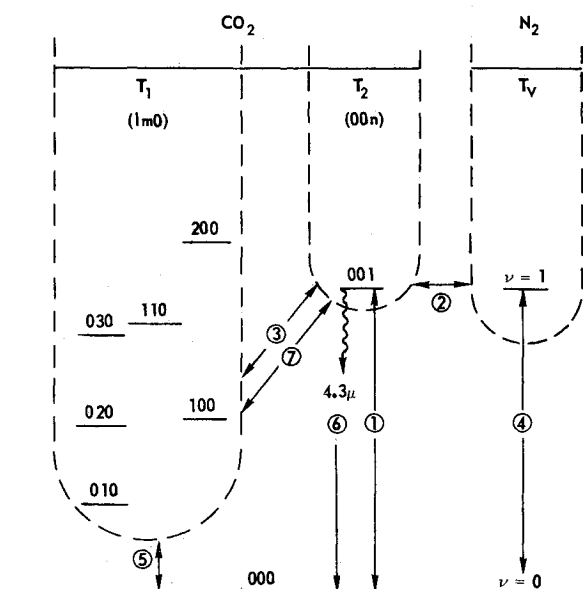


Fig. 3 CO₂-N₂ vibrational level diagram indicating important collision processes which are numbered in accordance with the text.

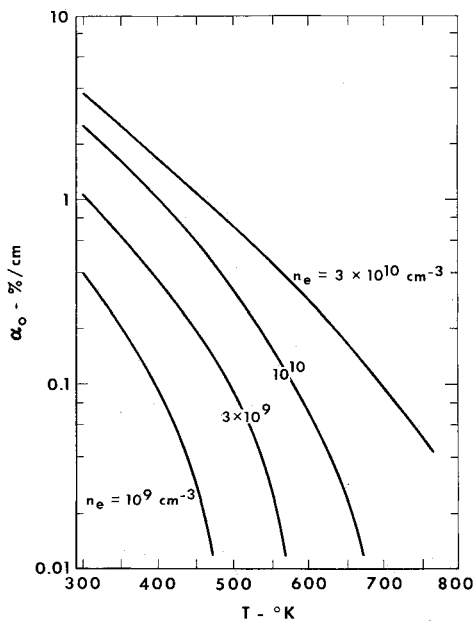


Fig. 5 Variation of small signal gain with electron density and gas temperature for a CO₂-N₂-He 0.1-0.1-0.8 laser mixture at a pressure of 10 torr.

and mixtures typical of CO₂ electric discharge laser operation. Shown in Fig. 5 is the computed variation of α_0 with electron density and gas temperature for a CO₂-N₂-He 0.1-0.1-0.8 mixture with a $\bar{u}_e$ value of 1.5 ev. At fixed gas temperature, the net rate of energy deposition in the CO₂ asymmetric stretch mode increases as the electron density is increased, resulting in an increase in the upper laser level population. For the values of electron density considered here, the population of the lower laser level is not appreciably enhanced as the electron density is increased due to the relatively large value of the atom-molecule relaxation rate for process 5 (Fig. 3). For these reasons the small signal gain increases with increasing electron density for constant gas temperature (Fig. 5). Increasing the value of gas temperature has a deleterious influence on the values of all the quantities of importance in determining small signal gain. 1) Since the magnitude of T_1 is close to T , increasing the latter results in an increase in the population in the lower laser level; 2) the rate constant for collisional relaxation of the upper laser level (process 3) increases with increasing gas temperature, resulting in a decrease in the population of the upper laser level; and 3) the value of the cross section for stimulated emission decreases with increasing temperature as is indicated by the data of Fig. 4. Therefore, for a fixed electron density, the value of α_0 decreases rapidly with increasing gas temperature.

The dependence of saturation intensity on electron density and gas temperature is shown in Fig. 6. Again the influence of these quantities on the various collisional rates manifests itself in the variation of saturation intensity. The saturation intensity is expected to increase as the cumulative excitation and de-excitation rates associated with the various collisional processes influencing the CO₂ asymmetric stretch mode increase.¹⁵ Therefore, the saturation intensity increases with both electron density and gas temperature as is indicated by the data of Fig. 6.

The sensitivity of α_0 and I_s to variations in electron density and gas temperature is reflected by the dependence of optical power density $\alpha_0 I_s$ on these quantities as is indicated by the data of Fig. 7. These data show that the strong detrimental effect of rising gas temperature on gain causes the optical power density to decrease in spite of the increase in saturation intensity. Therefore, it becomes clear that in most situations, the existence of a maximum in CO₂ laser output power as discharge current (electron density) is increased is a reflection of the accompanying rise of gas temperature. Even for high electron densities the available optical power density is reduced to nearly zero if the gas temperature

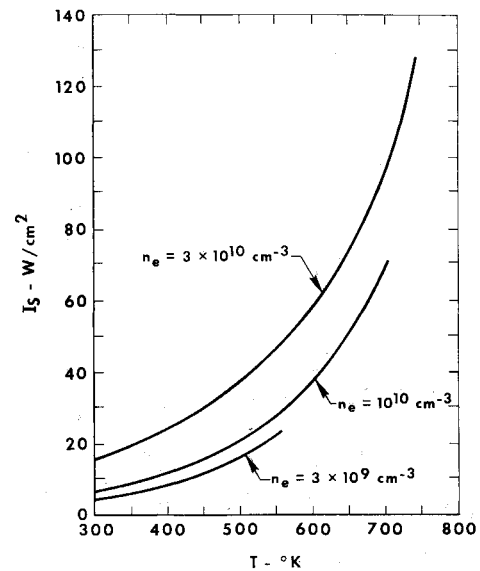


Fig. 6 Variation of 10.6 μ saturation intensity with electron density and gas temperature for the conditions of Fig. 5.

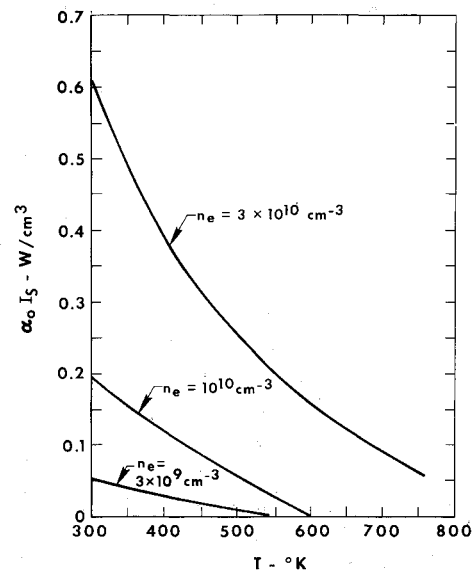


Fig. 7 Variation of maximum optical power density with electron density and gas temperature for the conditions of Fig. 5.

reaches approximately 700°K. In fact, the gain falls below practically useful levels in most CO₂ laser applications if the gas temperature exceeds about 600°K. For these reasons gas temperature is one of the most important parameters in determining potential power output of the CO₂ laser and therefore a strict control of gas temperature is a requisite for achievement of optimum laser performance. These considerations led to the development of the highly successful convection cooled CO₂ lasers¹⁶⁻¹⁸ which permit discharge power density levels substantially higher than conventional discharges while maintaining the gas temperature within the constraints discussed previously.

IV. Discharge Processes

Given a suitable CO₂ laser mixture, the electric discharge E/N ratio must be optimized to provide electrons with an energy distribution appropriate for efficient excitation of the key laser levels, while providing sufficient ionization to replenish the charged particles lost by ambipolar diffusion and/or recombination. In addition, the results of the previous section indicate that laser power density increases for a given mixture and E/N value with increasing electron density and decreasing gas temperature.

However, since electron density and gas temperature are not independent in an electric discharge, a detailed knowledge of local discharge properties is required for quantitative application of analytical results to laboratory situations. In order to study the influence of discharge plasma characteristics on laser properties, a modified Schottky analysis¹⁹ of a cylindrical positive column of a conventional, wall-dominated CO₂ laser discharge was developed. This analysis provides insight into the plasma processes occurring in the CO₂ laser discharges and supplies a means of confirming experimentally many of the analytic results discussed in previous sections.

Positive Column Analysis

A positive column analysis similar to that described by Ecker and Zoller²⁰ has been developed to calculate the radial variation of electron density and gas temperature in a cylindrical wall-dominated electric discharge laser. Required information for this calculation includes electron transport data such as the mobility and diffusion coefficients and the various electron-molecule ionization rates. These data were obtained using the computed electron energy distributions (Sec. II) appropriate to the gas mixture and specific E/N value. In addition, heavy particle $V-T$ processes are sufficiently fast in CO₂ laser mixtures so that the principal plasma energy loss process for this situation is thermal conduction to the tube walls. Therefore, knowledge of the temperature dependent thermal conductivity of the gas mixture is required. Further, the identity and mobility of the dominant positive ion must be known.

A detailed study of the numerous possible ionization, recombination, and charge exchange processes in CO₂ laser mixtures indicates that direct electron impact ionization of CO₂ from the ground state dominates the charged particle production process in mixtures containing approximately equal quantities of CO₂ and N₂. Multistage ionization processes involving N₂ metastable levels will likely become important in laser mixtures containing substantially more N₂ than CO₂, however. Although CO₂⁺ is readily produced by electron collisions, O₂⁺ is likely to be the dominant positive ion in CO₂ laser discharges. The O₂⁺ arises from electron impact dissociation of CO₂ in the discharge²¹ and, for most discharge operating conditions, is available in sufficient quantity to become positively charged as a result of charge exchange collisions with CO₂⁺ (Ref. 22). The positive ion remains O₂⁺ unless a molecule and/or atom with a lower ionization potential becomes available either through addition to the discharge or by means of discharge induced chemical reactions within the mixture. Positive ions are lost by wall recombination with electrons in the diffusion dominated CO₂ laser while dissociative recombination²³ is likely to be the dominant charged particle loss process in volume dominated laser systems.

Assuming that single-step ionization of CO₂ dominates ion production, that O₂⁺ is the dominant positive ion, and that diffusion of ions to the tube walls is the dominant ion loss process, the plasma energy equation and the charged particle continuity equations have been solved simultaneously for a CO₂-N₂-He (0.1-0.1-0.8) mixture leading to determination of the gas temperature and electron density profiles. Knowledge of the radial variations of n_e and T permits calculation of other important discharge properties including axial and radial currents, radial electric fields, and local values of E/N and $\bar{u}_e$. The results and implications of this analysis are discussed in subsequent paragraphs.

Variation of Discharge Properties

For conditions encountered in the wall dominated CO₂ electric discharge laser the radial variation of electron density was found to be nearly parabolic in form (the zero order Bessel function results only if T is nearly independent of radius). Computed values of the electron density on the discharge axis varied between 10^9 and 3×10^{10} electrons/cm³ depending on discharge current. Gas temperature also varied radially in a nearly parabolic manner while values on axis ranged between 400°K and 700°K over

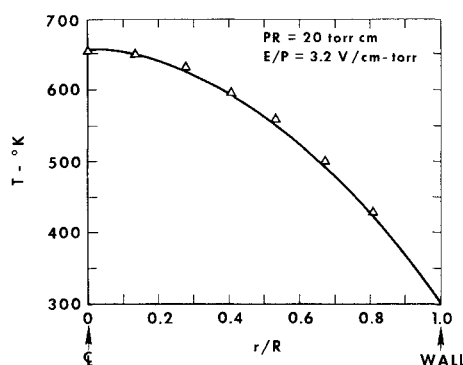


Fig. 8 Computed and measured radial gas temperature profiles for a wall-dominated CO₂ laser discharge with a radius of 1.9 cm. The total pressure of the CO₂-N₂-He 0.1-0.1-0.8 mixture was 10.5 torr.

the normal range of operating conditions. A typical radial gas temperature profile is shown in Fig. 8 along with experimentally measured values obtained using an electrically floating thermocouple. Very good agreement between analytically and experimentally determined gas temperature values is obtained throughout the full range of operating conditions, thus verifying that the thermal conduction model is valid for the wall dominated CO₂ laser in which He is present in quantity significantly greater than either CO₂ or N₂.

Using the calculated values of electron density and gas temperature, the relationship between discharge current and electric field was determined for direct comparison with experimental measurements. The discharge characteristics are most conveniently expressed in terms of the current-pressure product and E/P ratio as indicated in Fig. 9. Again, generally good agreement is obtained, the difference between measured and computed

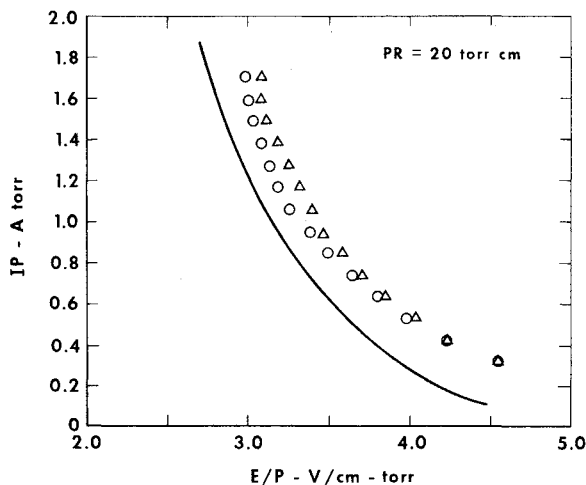


Fig. 9 Computed and measured variation of discharge current-voltage characteristics for the conditions of Fig. 8. The different symbols correspond to different experimental runs.

characteristics being well within the combined uncertainties of the experiment and the analysis. The negative resistance $IP-E/P$ characteristic is typical of discharges of this type and is due to the influence of changing gas temperature on the E/N ratio of the discharge and hence on the ionization rate which is exponentially dependent on E/N .

The satisfactory agreement between calculated and measured discharge characteristics (Fig. 9) indicates that the essential physical mechanisms have been included in the positive column analysis. In particular, the rationale leading to the assumption of a single step electron-CO₂ ionization process appears to have been verified. If, for example, multistage ionization processes made a substantial contribution to the ionization rate, measured

E/P values would be less than theoretical values for constant discharge current. Although the difference between theory and experiment is not considered significant, the data of Fig. 9 suggest that the single-step production process as treated in the present work is more than sufficient to provide the necessary ionization.

V. Radial Gain Profiles

Since plasma electron density and gas temperature are strong functions of radial position in CO_2 laser columns, the results of Sec. III (Figs. 5 and 6) suggest corresponding variations in laser optical properties. In order to investigate this likelihood, the radial variation of small signal gain was computed in a self-consistent fashion by combining the results of the analyses described in Sec. II-IV. In addition, for the purpose of comparison, values of α_0 at several radial positions were measured using a technique similar to that described by Smith and McCoy.²⁴ Shown in Fig. 10 is the radial variation of small signal gain so determined.²⁵ At very low current levels heating of the gas is minimal, ($N_{001} \gg N_{100}$), and the small signal gain should exhibit

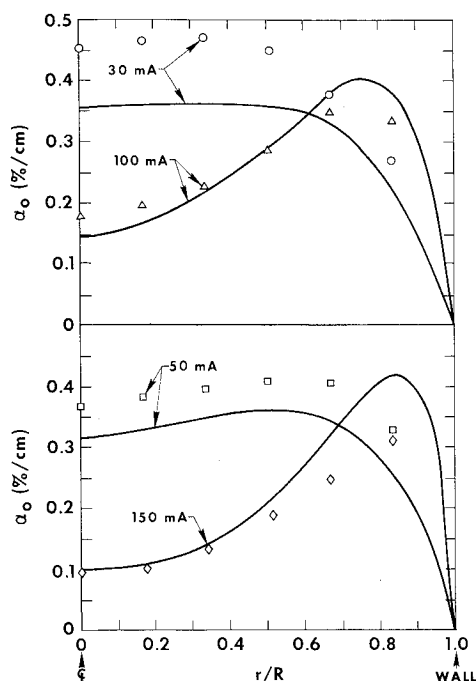


Fig. 10 Computed and measured small signal gain profile as a function of discharge current for the conditions of Fig. 8.

a nearly parabolic radial variation reflecting the importance of the electron density profile in determining the upper laser level population, N_{001} . However, with increasing discharge current the strong influence of gas temperature (Figs. 5 and 8) on the lower laser population becomes important, ($N_{001} > N_{100}$). For the present experiment this situation is manifest at the 30 ma current level for which the potential increase of the population inversion due to an increase in electron density is offset by the detrimental effect of an increase of axial gas temperature. This results in a small signal gain profile which is nearly independent of radius except near the wall. Further increases in current accentuate this trend. For current levels above approximately 50 ma, the small signal gain on center line is less than at points closer to the tube walls, indicative of the very strong influence of local gas temperature on the population of the CO_2 (100) level, on the collisional relaxation rate of the (001) level, and on the cross section for stimulated emission. In fact, the importance of rising gas temperature is so great that the magnitude of the axial gain is found to continuously decline with increasing current despite the concomitant increase in electron density and therefore discharge power density. The generally good qualitative and quantitative

agreement between theory and experiment exemplified by the data of Fig. 10 is taken as verification of the validity of the various analytic modeling techniques discussed previously.

VI. Discussion

The results obtained from the analyses of the electron and molecular kinetics and the positive column analysis have provided considerable insight into the physics of CO_2 laser discharges. The details discussed in the previous sections for a typical CO_2 - N_2 -He mixture illustrate the techniques and physical models required to analyze a wall-dominated CO_2 electric discharge laser column. The findings of these studies are rather general, however. The calculated results of the electron and molecular kinetics analyses are for the most part independent of discharge configuration, depending only on the local discharge properties, i.e., n_e , T , E/N , and the gas mixture, and are therefore applicable to a broad class of electrically excited CO_2 lasers. As illustrated in part by Figs. 2, 5, 6, and 7, there is a range of each of these parameters for which adequate gain and efficient laser excitation are possible. In addition, the strong detrimental influence of an elevated gas temperature on laser performance was amply illustrated by the results of Secs. III and V. The nature of this information permits generalization of the findings of the present analyses for application to more practical discharge conditions and/or configurations.

Since the gas temperature appears to be one of the most important parameters affecting the laser performance, means of controlling gas temperature should provide increased laser power and efficiency. A technique which accomplishes this is convective cooling.¹⁶⁻¹⁸ Instead of providing heat removal by thermal conduction to the discharge tube walls, heat is removed convectively by the flowing gas mixture.¹⁶⁻¹⁸ By specifying a reasonable temperature rise and gas pressure, the required flow velocity of the gas can be readily calculated. In rectangular channels, gas flow perpendicular to the discharge or optical axes of the laser provides minimum temperature elevation for minimum flow velocity. The resulting permissible increase in discharge power density leads to laser output power at the kw level¹⁶⁻¹⁸ with modest flow velocities on the order of 100 m/sec. A further advantage of convective cooling is that the spatial variation in gas temperature is minimized providing a laser medium of nearly uniform gain. Moreover, convectively cooled CO_2 lasers may be built as closed-cycle devices thus conserving the laser gas mixture.

For the reasons elaborated in Sec. III, maintenance of the gain at an acceptable level places a constraint on the maximum tolerable gas translational temperature in CO_2 lasers, i.e., $T < 600$ – 700°K . However, this limit is seldom reached even in high power convection dominated lasers due to the onset of discharge instability leading to local streamering and/or transient arcs which seriously impede laser performance. This phenomenon may be attributed, at least in part, to chemical reactions occurring in the discharge which significantly alter the composition of the laser mixture.²⁶ In conventional CO_2 lasers the direct dissociation of CO_2 by electron impact has been found to be very important²¹ in that CO_2 dissociation results not only in a loss of CO_2 , but can adversely affect the electron energy distribution and molecular kinetics processes as well. Calculations³ and experiments²¹ indicate that a decrease in laser performance accompanies fractional CO_2 dissociations exceeding approximately 10%. Even a small fractional dissociation of CO_2 results in changes in the dominant positive ion in many discharges from CO_2^+ to O_2^+ . Since the effective dissociation time is on the order of 0.1–1.0 sec, for the flow velocities usually associated with convectively cooled CO_2 lasers, CO_2 dissociation is usually less than a few percent. However, if the gas mixture is reused as in a closed-cycle laser, the dissociation of CO_2 will proceed to the equilibrium value determined by discharge conditions.

A time scale comparable to the typical gas residence time in convection-dominated CO_2 lasers ($\sim 10^{-2}$ sec) is sufficient to produce other significant changes in the composition of mixtures composed initially of CO_2 , N_2 , and He. The dissociation of CO_2 , and to a far lesser extent N_2 , produces CO, O, and N which subsequently leads to the formation of quantities of NO, N_2O , and

NO₂ which reach fractional concentrations on the order of parts per million.²⁶ In closed-cycle lasers these molecules accumulate to densities such that the dominant positive and negative ions are directly related to the minority species. These impurity species lead to a host of complex positive and negative ions which may have densities significantly in excess of that of the electrons. Of particular importance is evidence suggesting that appreciable negative ion concentrations may contribute to the onset and development of discharge instabilities which seriously deteriorate discharge performance at high pressure (> 10 torr) and for high power densities.

In order to circumvent discharge stability and plasma chemistry problems, recent attention has been directed toward development of techniques whereby the principal ionization processes can be decoupled from the discharge processes, i.e., from E/N . Such efforts have led to implementation of the electron beam²⁷ sustained CO₂ laser discharge in which ionization is provided uniformly throughout the laser volume with a high energy (~100 keV) electron beam thereby permitting independent adjustment of E/N to provide optimum vibrational excitation (Fig. 2). In addition to improving vibrational excitation efficiency, this scheme permits operation of convection dominated discharges at substantially higher power densities by changing the threshold for onset of discharge instability. The use of auxiliary rf electric fields to augment d.c. CO₂ laser discharges has also proved an effective means of stabilizing the discharge under high-power conditions.²⁸ In addition, the minimization of local thermal inhomogeneities, which contribute to discharge instability, by judicious introduction of small scale turbulence has proved successful in some circumstances.²⁹ However, despite the success that has been achieved with various methods of discharge stabilization, at present the role of plasma chemistry and other fundamental factors influencing laser discharge stability is only partially understood. Since discharge instability may represent a serious practical limitation to the development of high power electrically excited molecular lasers, additional research in this area is clearly required.

VII. Summary

This paper analyzes the physics of CO₂ electric discharge lasers and serves to illuminate the fundamental physical processes influencing CO₂ laser performance. The results of recent studies of electronic, molecular, and plasma processes have been reviewed and their significance as regards CO₂ laser operation are discussed. These studies demonstrate the significant influence of electron-molecule collision phenomena on the electron distribution function in CO₂ lasers, and thus upon the source of excitation of the relevant CO₂ and N₂ vibrational levels. In addition, the results of examination of discharge ionization kinetics, ion charge exchange kinetics, and charged particle loss processes, suggest that for many CO₂ laser conditions electron impact ionization of CO₂ is the dominant ionization process, with CO₂⁺ and/or O₂⁺ the most likely surviving ionic species. Further, the analyses discussed herein illustrate the dependence of small signal gain and saturation intensity on CO₂ discharge properties. These data are of primary importance, indicating that merely increasing electrical input power to a CO₂ laser discharge cannot be expected to increase laser power if accompanied by an increase in gas temperature to a level exceeding approximately 600°K. Gas temperature is therefore seen to be a fundamental limiting parameter which significantly influences laser performance. However, many high power CO₂ laser systems are found to operate well below the thermal limit as a result of discharge instability, the onset of which appears to be strongly influenced by the presence of stable negative ions formed from discharge produced impurity species. For this reason, further investigation of the factors influencing discharge stability, particularly discharge induced chemical reactions, is of considerable importance.

References

- ¹ Sobolev, N. N. and Sokovikov, V. V., "CO₂ Lasers," *Soviet Physics Uspekhi*, Vol. 10, No. 2, Sept.-Oct. 1967, pp. 153-170.
- ² Nighan, W. L. and Bennett, John H., "Electron Energy Distribution Functions and Vibrational Excitation Rates in CO₂ Laser Mixtures," *Applied Physics Letters*, Vol. 14, No. 8, April 1969, pp. 240-243.
- ³ Nighan, W. L., "Effect of Molecular Dissociation and Vibrational Excitation on Electron Energy Transfer in CO₂ Laser Plasmas," *Applied Physics Letters*, Vol. 15, No. 11, Dec. 1969, pp. 355-357.
- ⁴ Nighan, W. L., "Electron Energy Distributions and Collision Rates in Electrically Excited N₂, CO, and CO₂," *Physical Review (A)*, Vol. 2, No. 5, Nov. 1970, pp. 1989-2000.
- ⁵ Fowler, M. C., "A Quantitative Analysis of the Dependence of CO₂ Laser Performance on Electric Discharge Properties," *Applied Physics Letters*, Vol. 18, No. 5, March 1970, pp. 175-178.
- ⁶ Engelhardt, A. G., Phelps, A. V., and Risk, C. G., "Determination of Momentum Transfer and Inelastic Collision Cross Sections for Electrons in Nitrogen Using Transport Coefficients," *Physical Review*, Vol. 135, No. 6A, Sept. 1964, pp. A1566-A1574.
- ⁷ Hake, R. D., Jr. and Phelps, A. V., "Momentum Transfer and Inelastic Collision Cross Sections for Electrons in O₂, CO, and CO₂," *Physical Review*, Vol. 158, No. 1, June 1967, pp. 70-84.
- ⁸ Phelps, A. V., private communication, 1971, Joint Inst. for Laboratory Astrophysics.
- ⁹ Taylor, R. L. and Bitterman, S., "Survey of Vibrational Relaxation Data for Processes Important in the CO₂-N₂ Laser System," *Review of Modern Physics*, Vol. 41, No. 1, Jan. 1969, pp. 26-47.
- ¹⁰ Gordietz, B. F., Sobolev, N. N., Sokovikov, V. V., and Shelepin, L. A., "Population Inversion of Vibrational Levels in CO₂ Lasers," *IEEE Journal of Quantum Electronics*, Vol. QE 4, No. 11, Nov. 1968, pp. 796-802.
- ¹¹ Moore, C. B., Wood, R. E., Hu, B.-L., and Yardley, J. T., "Vibrational Energy Transfer in CO₂ Lasers," *Journal of Chemical Physics*, Vol. 46, No. 11, June 1967, pp. 4222-4231.
- ¹² Stephenson, J. C., Wood, R. E., and Moore, C. B., "Near-Resonant Energy Transfer Between Infrared-Active Vibrations," *Journal of Chemical Physics*, Vol. 48, No. 10, May 1968, pp. 4790-4791.
- ¹³ Herzberg, Gerhard, *Molecular Spectra and Molecular Structure, II Infrared and Raman Spectra of Polyatomic Molecules*, D. Van Nostrand Co., Princeton, N.J., 1968, pp. 215-218.
- ¹⁴ Herzfeld, K. F. and Litovitz, T. A., *Absorption and Dispersion of Ultrasonic Waves*, Academic Press, New York, 1959, p. 86.
- ¹⁵ Christensen, C. P., Freed, C., and Haus, H. A., "Gain, Saturation and Diffusion in CO₂ Lasers," *IEEE Journal of Quantum Electronics*, Vol. QE-5, No. 6, June 1969, pp. 276-283.
- ¹⁶ Tiffany, W. B., Targ, R., and Foster, J. D., "Kilowatt CO₂ Gas-Transport Laser," *Applied Physics Letters*, Vol. 15, No. 3, Aug. 1969, pp. 91-93.
- ¹⁷ Brown, C. O., "Plasma Properties of an Electric Discharge Convection Laser," *Bulletin of the American Physical Society*, Vol. 16, No. 2, Feb. 1971, p. 215.
- ¹⁸ Eckbreth, A. C. and Davis, J. W., "Cross-Beam Electric-Discharge Convection Laser," *Applied Physics Letters*, Vol. 19, No. 4, Aug. 1971, pp. 101-103.
- ¹⁹ Francis, Gordon, "The Glow Discharge at Low Pressure," *Encyclopedia of Physics*, Vol. 22, S. Flugge, ed., Springer-Verlag, Berlin, 1956, p. 114.
- ²⁰ Ecker, G. and Zoller, O., "Thermally Inhomogeneous Plasma Column," *The Physics of Fluids*, Vol. 7, No. 12, Dec. 1964, pp. 1996-2000.
- ²¹ Wiegand, W. J., Fowler, M. C., and Benda, J. A., "Carbon Monoxide Formation in CO₂ Lasers," *Applied Physics Letters*, Vol. 16, No. 6, March 1970, pp. 237-239.
- ²² Norton, R. B., Ferguson, E. E., Fehsenfeld, F. C., and Schmeltekopf, A. L., "Ion-Neutral Reactions in the Martian Ionosphere," *Planetary and Space Sciences*, Vol. 14, No. 10, Oct. 1966, pp. 969-978.
- ²³ Mehr, F. J. and Biondi, M. A., "Electron Temperature Dependence of Recombination of O₂⁺ and N₂⁺ Ion with Electrons," *Physical Review*, Vol. 181, No. 1, May 1969, pp. 264-271.
- ²⁴ Smith, D. C. and McCoy, J. C., "Effects of Diffusion on the Saturation Intensity of a CO₂ Laser," *Applied Physics Letters*, Vol. 15, No. 1, Jan. 1969, pp. 1-3.
- ²⁵ Wiegand, W. J., Fowler, M. C., and Benda, J. A., "Influence of Discharge Properties on CO₂ Laser Gain," *Applied Physics Letters*, Vol. 18, No. 9, May 1971, pp. 265-267.
- ²⁶ Wiegand, W. J., "The Influence of Plasma-Generated Impurities on Molecular Gas Discharges," *Bulletin of the American Physical Society*, Vol. 17, No. 3, March 1972.
- ²⁷ Fenstermacher, C. A., Nutter, M. J., Rink, J. P., and Boyer, K., "Electron Beam Initiation of Large Volume Electrical Discharges in CO₂ Laser Media," *Bulletin of the American Physical Society*, Vol. 16, No. 1, Jan. 1971, p. 42.

²⁸ Haas, R. A. and Guckel, G., "Stability of Electric Discharges in Flowing Molecular Gases," *Bulletin of the American Physical Society*, Vol. 16, No. 11, Nov. 1971, p. 1283.

²⁹ Hill, A. E., "Uniform Electrical Excitation of Large-Volume High-Pressure Near-Sonic CO₂-N₂-He Flowstream," *Applied Physics Letters*, Vol. 18, No. 5, March 1971, pp. 194-197.

APRIL 1972

AIAA JOURNAL

VOL. 10, NO. 4

Analysis of an Electric Discharge CO₂ Mixing Laser

H. A. HASSAN* AND J. W. BORDEAUX†
North Carolina State University, Raleigh, N.C.

A one-dimensional model of an electric discharge CO₂ mixing laser is presented. The model takes into consideration chemical, vibrational, and thermal nonequilibrium effects. Starting from the entrance of the glow discharge duct, the conservation equations for the various constituents are integrated through the glow discharge and mixing regions. The effects of pressure, mass flow rates of the various species, and current density are discussed in detail. It is shown that, for given pressure and temperature at the entrance, the gain coefficient and power output increase with an increase in CO₂ flow rate; also, there is a current density, a He flow rate and an N₂ flow rate at which the gain coefficient is constant.

Introduction

RECENT interest in high-flow electric discharge CO₂ laser systems results from their continuous high-power capabilities and high efficiencies. Although the inversion mechanism in CO₂ molecules is well understood, there is a need to formulate a self-consistent model which takes into consideration the chemical, vibrational, and thermal nonequilibrium effects and thus serves as a basis for optimizing the laser parameters of interest.

The problem considered here is that of the electric discharge mixing laser system shown in Fig. 1. In this system, N₂ and He are premixed in an upstream stagnation chamber and the mixture is excited by a rectangular glow discharge. The CO₂ is injected downstream of the anode and upstream of the optical axis.

The model presented here treats molecules in different quantum states as different species and employs the conservation of species equations, the over-all momentum and energy equations, and an

electron energy equation to describe the laser system. A survey of the various relaxation processes involving heavy particles and available rates was given in Ref. 1. The important reactions indicated in Ref. 1, together with appropriate electron-heavy particle collisions serve as the basis of the kinetics in this work. Thus, the three-level approximation for the energy levels of CO₂ (Refs. 2 and 3) is not employed.

The one-dimensional approximation is employed; this implies that the laser is of the gas transport type. The resulting equations are integrated starting from the entrance of the glow discharge duct through the glow discharge and mixing regions. As a result of this integration, one can calculate, as a function of the distance, the gain coefficient, power output, number densities of the various levels, gas and electron temperatures, velocity, and pressure. The effects of pressure, mass flow rates of the various species, and current density on the gain coefficient and power output are discussed in detail.

Analysis

Treating molecules in different quantum states as different species, the governing equations are then the conservation of species equations, the over-all momentum and energy equations, and an electron energy equation.⁴ The conservation of species equations can be written as

$$\partial \rho_i / \partial t + \nabla \cdot (\rho_i \mathbf{u}_i) = m_i R_i \equiv w_i \quad (1)$$

where, for species i , ρ_i is the density, m_i is the particle mass, $\mathbf{u}_i$ is the velocity, and R_i is the rate of production of species i . As a result of conservation of mass, summation of Eq. (1) with respect to i yields

$$\partial \rho / \partial t + \nabla \cdot (\rho \mathbf{u}) = 0 \quad (2)$$

where $\rho = \sum \rho_i$ is the mean density and $\mathbf{u} = (\sum \rho_i \mathbf{u}_i) / \rho$ is the mean velocity of the entire system. Letting

$$\mathbf{u}_i = \mathbf{u} + \mathbf{V}_i, \quad Y_i = \rho_i / \rho \quad (3)$$

where $\mathbf{V}_i$ is the diffusion velocity, substituting into Eq. (1) and simplifying the resulting expression by using Eq. (2), one obtains

$$\rho (\partial Y_i / \partial t) + \rho \mathbf{u} \cdot \nabla Y_i + \nabla \cdot (Y_i \mathbf{V}_i) = w_i \quad (4)$$

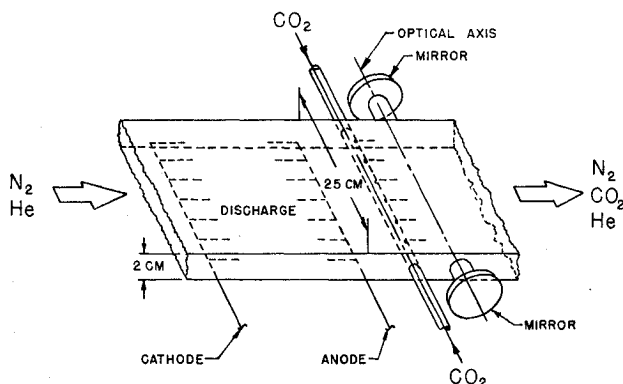


Fig. 1 Schematic of an electric discharge mixing laser.

Presented as Paper 71-66 at the AIAA 9th Aerospace Sciences Meeting, New York, January 25-27, 1971; submitted February 1, 1971; revision received September 29, 1971. Supported, in part, by NASA Grant NGR 34-002-115 and NSF Grant GY-7632.

Index categories: Lasers; Plasma Dynamics and MHD.

* Professor of Mechanical and Aerospace Engineering. Associate Fellow AIAA.

† Research Assistant.