QUASISTATIONARY MODE OF CO_2-LASER EXCITATION BY A NONINDEPENDENT DISCHARGE

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Dynamics of the operation of a CO_2-laser during excitation by a nonindependent discharge is investigated within the framework of a comparatively simple theoretical model.

A nonindependent electrical discharge in a gas is one of the most prospective methods of pumping CO_2-lasers. Under the conditions of such a discharge the mean electron energy and their concentration are independent quantities, which permits realization of the most favorable excitation mode for the upper lasing level. Moreover, an external ionization source facilitates realization of a homogeneous discharge at high pressures of the working medium. An additional argument in favor of using a combined discharge is the fact that optimal values of the electric field intensity, in the sense of laser pumping efficiency, turn out to be somewhat lower than that required to sustain an independent discharge in typical laser mixtures, whereupon the system as a whole becomes less responsive to the development of an overheating-ionization instability. As an illustration of the practical utilization of a discharge of the kind mentioned to pump a CO_2-laser, the papers [1, 2] can be mentioned, in which the volume ionization of the discharge gap is realized by using an electron beam and a sequence of short high-voltage pulses, respectively.

In the presence of the possibility for an independent variation in the electron density n which is characteristic for a nonindependent discharge, the question of the dependence of the lasing system parameters on the value of n for a fixed value of the parameter E/N of the main discharge becomes important in principle and in practice. An optimal value of the electron concentration n* corresponding to the maximum value of the lasing efficiency should exist for each given composition and pressure of the working mixture. Taking into account that the electric field intensity of the main discharge is selected from the condition of maximum efficiency of exciting the upper level of the lasing transition, it can be asserted that the quantity n* will govern the corresponding "optimal" values of all the main parameters of a laser with nonindependent discharge. The question of the possibility of realizing a quasistationary generation mode under the conditions of a nonindependent discharge is also of substantial interest. Variation of the quantity n can be used to control the duration and intensity of a laser radiation pulse.

In conformity with the above, the need for a quantitative analysis of the dependence of the properties of the lasing system on the magnitude of the electron density under combined discharge conditions is evident. An attempt is made in this paper at such an analysis for a comparatively simple general formulation of the problem. The following additional assumptions about the nature of the discharge were hence used.

1. The initial electron concentration originates instantaneously at the time $t = 0$ and enters the equation as a variable parameter. Variations with a quasistationary and pulse mode of ionization source operation were examined. In the former case, the relative electron concentration n/N is considered constant in time, while in the latter dissociative recombination is taken into account.

2. The electric field intensity during the discharge varies so that the quantity E/N remains constant. The value $E/N = 3 \cdot 10^{-16}$ V · cm^2 was used in specific computations. The appropriate value of the mean electron energy, calculated taking into account that the velocity distribution function is not Maxwellian, is about 2.5 eV [3, 4].

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TABLE 1

No.	Direct and reverse process	k^+ (→)	k^- (←)	Reference
1	$N_2+e \rightleftarrows N_2^*+e$	$k_{eN}=2{,}4\cdot10^{-8}$ cm$^3\cdot$sec^{-1}	$1{,}2\exp\left(-\frac{h\nu_4}{kT_e}\right)k^+$	[3, 4, 6]
2	$CO_2+e \rightleftarrows CO_2^*(001)+e$	$0{,}25\ k_{eN}$	$\exp\left(\frac{h\nu_3}{kT_e}\right)k^+$	[3]
3	$CO_2+e \rightleftarrows \begin{cases} CO_2^*(100)+e \\ CO_2^*(020)+e \end{cases}$	$0{,}5\ k_{eN}$	$\exp\left(\frac{h\nu_1}{kT_e}\right)k^+$	[3]
4	$CO_2+e \rightleftarrows CO_2^*(010)+e$	$0{,}2\ k_{eN}$	$\exp\left(h\nu_2/kT_e\right)k^+$	[3]
5	I: $N_2^*(n+1)+CO_2 \rightarrow N_2^*(n)+CO_2^*(001)$; $CO_2^*(00n)+N_2^*(1) \leftarrow CO_2^*(00n+1)+N_2$ II: $N_2^*(n+1)+CO_2 \rightarrow N_2^*(n)+CO_2^*(001)$; $CO_2+N_2^*(1) \leftarrow CO_2^*(001)+N_2$	$\left(0{,}2+\frac{2}{W}\right)10^5\ k_{eN}$	$\exp\left(\frac{\Delta_{34}}{kT}\right)k^+$	[9—11, 14]
6	$CO_2^*(001)+CO_2 \rightarrow \begin{cases} CO_2^*(030)+CO_2 \\ CO_2^*(110)+CO_2 \end{cases}$	$50\exp\left(16{,}3\ W^{-\frac{2}{3}}-35\ W^{-\frac{1}{3}}\right)k_{eN}$		[9, 11, 13—15]
7	$CO_2^*(001)+He \rightarrow \begin{cases} CO_2^*(030)+He \\ CO_2^*(110)+He \end{cases}$	$9{,}2\cdot10^{-4}\exp\left(-10\ W^{-1/3}\right)k_{eN}$		[9, 12, 14, 15]
8	I: $CO_2^*(0n+10)+He \rightleftarrows CO_2^*(0n0)+He$ II: $CO_2^*(010)+He \rightleftarrows CO_2+He$; $CO_2^*(020)+He \rightleftarrows CO_2^*(010)+He$	$2\cdot10^{-3}\exp\left(-6{,}1\ W^{-1/3}\right)k_{eN}$	$2\exp\left(-\frac{h\nu_2}{kT}\right)k^+$	[9, 14]
9	II: $CO_2^*(020)+CO_2 \rightleftarrows 2CO_2^*(010)$	$6{,}3\cdot10^{-4}\ W^{1/2}\ k_{eN}$	$0{,}5\ k^+$	[9]
10	Stimulated radiation $(001) \rightarrow (100)CO_2$			

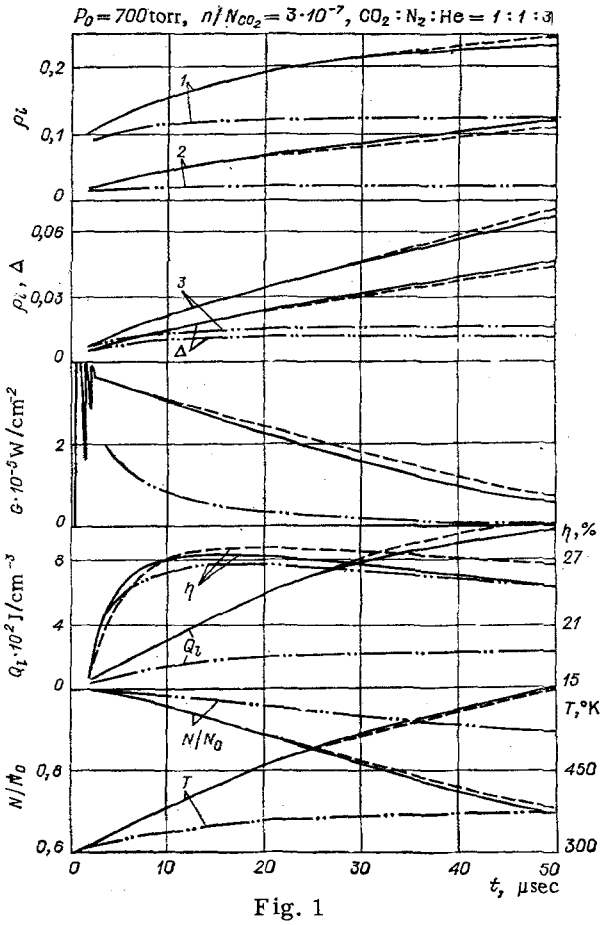

Fig. 1

3. The pumping intensity is sufficiently large; consequently, diffusion and heat-exchange processes can be neglected during the generation pulse whose duration is determined by the specific heat of the lasing mixture under the conditions mentioned.

4. The gas density in the discharge zone remains homogeneous during heating and expansion, by varying in time according to the law

$$N = N_0[T_0/T + (1 - T_0/T)e^{-t/\tau}], \tag{1}$$

where T is the gas temperature, $\tau \sim l/c_S$; l is the transverse dimension of the discharge, and c_S is the speed of sound. Elementary gasdynamic estimates show that the quantity τ can be taken equal to 1.5 l/c_S. Such an approximation permits significant simplification of the solution of the problem while conserving the most essential singularities of the phenomenon under consideration. In the limit cases of $t \ll \tau$ and $t \gg \tau$ the relationship (1) goes over into $N = N_0 = \text{const}$ and $N = N_0T_0/T$, respectively. The initial values of the gas temperature and density were $T_0 = 300°K$ and $N_0 \geq 3.54 \cdot 10^{18}$ cm^{-3} ($P_0 \geq 100$ torr) in the computations carried out.

Under the assumptions mentioned, the problem as a whole is nonstationary and solved by numerical integration of the system of kinetic equations for the populations of the N_2 and CO_2 vibrational levels jointly with the equations for the gas temperature and the electron concentration. The equation for the gas temperature in the discharge zone is written on the basis of the relationship $dQ/dt = c_V N dT/dt - kT dN/dt$ and, taking account of (1), becomes

$$\frac{dQ}{dt} = c_V N_0 T_0 \left[\frac{\gamma}{W} + \left(1 - \frac{\gamma}{W}\right)e^{-t/\tau}\right]\frac{dW}{dt} + \frac{N_0 k T_0}{\tau}(W - 1)e^{-t/\tau},$$

where Q is the energy within unit volume of the discharge because of V–T relaxation processes and elastic electron collisions with molecules, and $\gamma = c_p/c_V$; $W = T/T_0$; k is the Boltzmann constant. The power den-

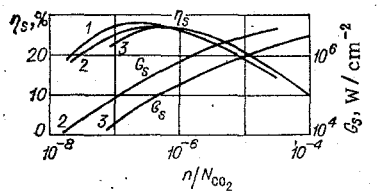

Fig. 2

sity liberated because of the elastic electron collisions is calculated by means of the formula $dQ_e/dt = nm^{1/2}(2\varepsilon)^{3/2}\sum_k \frac{\sigma_k N_k}{M_k}$, where ε and m are the mean electron energy and mass, respectively, M_k is the mass of molecules of the k-th component of the mixture, N_k are the partial concentrations, and σ_k are the mean sections of the elastic electron collisions with CO_2, N_2, and He, equal to $8.5 \cdot 10^{-16}$, $1.7 \cdot 10^{-15}$ and $5.1 \cdot 10^{-16}$ cm^2, respectively [5]. The role of the elastic loss channel is insignificant at moderate electron concentrations.

The equation for the electron concentration is

$$\frac{dn}{dt} = -\beta n^2 - n_0\left[\frac{1-e^{-t/\tau}}{W^2}\frac{dW}{dt} + \left(1-\frac{1}{W}\right)\frac{e^{-t/\tau}}{\tau}\right], \qquad (2)$$

where β is the coefficient of dissociative recombination. The values $\beta = 2 \cdot 10^{-7}$ cm^3/sec for a pulse ionization source and $\beta = 0$ for a stationary source (ionization because of the main electric field can be neglected under the conditions being considered). The second member in the right side of (2) describes the change in electron concentration because of the change in gas density in conformity with (1).

When writing the kinetic equations it is necessary to take account of the following circumstance. The sections of electron excitation of the lower vibrational levels of the N_2 molecules (down to v = 5) are quantities of the same order of magnitude [6]. This means that the effective population by a large set of nitrogen vibrational levels occurs under the conditions of an arbitrary discharge. Hence, the most suitable model to describe the role of N_2 in the system of kinetic equations is the model of a harmonic oscillator with a Boltzmann population of the vibrational levels and an appropriate vibrational temperature. As regards the vibrational levels of the fundamental CO_2 molecule modes, characteristic for them is an abrupt diminution in the electron-excitation section as the level number grows [7]. In this case population of the upper levels occurs mainly because of vibrational exchange processes within each mode competing with the sufficiently intensive stimulated radiation and vibrational–translational low-level deactivation. Hence, both a model based on the assumption of a Boltzmann population of levels within each of the modes interpreted as harmonic oscillators [8] and the model in which only levels directly related to the upper and lower lasing transition levels can be used to describe the kinetics of the vibrational exchange and relaxation of the CO_2 molecules.

Both the models mentioned were used in this paper to check out the machine computation program. It was established that both modifications yield close results. The main part of the last computations were carried out by the first modification. It should be noted that a comparison between the two models mentioned is of independent interest, since the second model applied to stationary problem conditions results in a simpler system of algebraic equations and hence turns out to be preferable.

Given in the Table 1 are the excitation, exchange, and deactivation reactions for the vibrational levels which were taken into account in writing the velocity equations, and empirical formulas are presented for the temperature dependences of the corresponding kinetic coefficients obtained by processing data from the survey [9] and the later papers [10-15]. The following notation was hence used: k^+, k^- rate constants of the forward and reverse processes; $h\nu_i$, T_i (i = 1, 2, 3, 4) the vibrational quantum energy and the vibrational temperature, respectively, for the modes (n00), (0n0), (00n)CO_2 and (v = n)N_2; $\Delta_{34} = h(\nu_3 - \nu_4)$; T_e is the electron temperature ($1.85 \cdot 10^{4}$°K). The numbers I and II indicate the reactions for the model with the harmonic oscillators and the combination model with separate CO_2 molecule levels, respectively. In both cases only single-quantum transitions were taken into account in the exchange and relaxation processes. It was assumed that the section magnitudes measured experimentally refer to transitions between the low levels, and the usual relationship $k_{n+1,n} = (n+1)k_{1,0}$ was used to evaluate transition sections of the type $(n+1) \rightarrow (n)$.

Let us briefly examine the main singularities of the reactions used in the computations.

1. The velocity of transferring excitation from N_2 to CO_2 in model II [see process (5) in the Table 1] turns out to be explicitly exaggerated. As checking computations showed, this difference is basic for the two models considered.

2. Processes reverse to (6) and (7), were not taken into account, since the velocities of these reactions are small compared to the others governing the population of the appropriate levels. The forward processes are essential for threshold generation and for small electron concentrations.

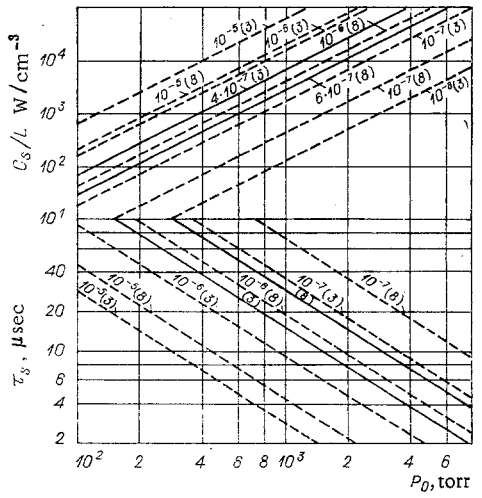

Fig. 3

3. The process (8) corresponds to the main V–T relaxation channel in the computations conducted. Single-quantum transitions from the first and second levels of the flexible CO_2 mode were hence taken into account in the model with the separate levels. The role of water vapor admixtures was not taken into account. Simple estimates show that the optimal admixture of H_2O vapor increases the velocity of deactivation of the lower lasing level 1.5-2-fold.

4. Equality of the vibrational temperatures of the flexible and symmetric CO_2 modes was assumed in the model with the harmonic oscillators because of the presence of Fermi resonance between the (100) and (020) levels. The fast "decay" reaction of $CO_2(020)$, $2CO_2(010)$ was taken into account in the model with separate levels.

5. The temperature dependence of the inelastic molecular collision cross section, accompanied by the change in the rotational quantum number [4, 16], was taken into account in the expression for the lasing transition. This yields a more abrupt change in the gain coefficient with temperature than the classical model with collisionally broadened lines. The general relationship for the gain coefficient can be written as $\alpha = g(T)\alpha_c$, where g(T) describes the mentioned additional temperature dependence, and α_c is the gain coefficient calculated under the assumption of constancy of the gas-kinetic sections. A formula for g(T) has been obtained as a result of processing the data from [4]:

$$g = 1.635\, W^{-2}\exp(-0.49/W).$$

The contribution from Doppler broadening in the line shape can be neglected in the $p \geq 100$-torr-pressure range. The quantity α_c at the center of the collisionally broadened line P, the branch of the lasing transition, is determined by the relationship

$$\alpha_c = \frac{A\lambda^2 (2j-1)\, hcB}{2\pi^2 \Delta\nu_c kT} \exp\left[-\frac{hcB}{kT} j(j-1)\right](n_2 - n_1 e^{-2j\frac{hcB}{kT}}), \qquad (3)$$

where $A = 0.165$ sec^{-1} [17, 18] is the probability of spontaneous radiation, $\lambda = 1.06 \cdot 10^{-3}$ cm is the lasing transition wavelength, j is the number of the rotational level of the lower vibrational state, $B \simeq 0.39$ cm^{-1} is a rotational constant, n_2, n_1 are populations of the CO_2 (001) and (100) levels, $\Delta\nu_c$ is the width at the half-altitude of the line shape, h is the Planck constant, and c is the speed of light (it is assumed that the population of the rotational levels is a Boltzmann distribution with temperature T). For a gas mixture with the partial concentrations N_i $\Delta\nu_c$ is calculated by the formula $\Delta\nu_c = \sum_i \Delta\nu_i N_i$, where $\Delta\nu_i$ is the specific (per particle) half-width. The values of $\Delta\nu_i$ for the CO_2–CO_2, CO_2–N_2, CO_2–He collisions were taken from [18, 19]. The computations were carried out for the line P (20) of the lasing transition. Substitution of the appropriate numerical values into (3) yields

$$\alpha_c\,(cM^{-1}) = \frac{6.78 W^{-3.2} e^{-\frac{0.71}{W}}}{(1.8N_c + 1.7N_N + 1.3N_H)}\,(n_2 - n_1 e^{-7.5\cdot 10^{-2}/W}).$$

For simplicity it was assumed that the gain coefficient is identical over the cavity volume and the radiation losses are associated only with the flux through the output mirror. The threshold generation conditions are determined by the customary relationship $2\alpha L = -\ln R$, where L is the length of the discharge and the cavity, and R is the coefficient of reflection of the output mirror.

The equation for the radiation density ρ in the cavity is $d\rho/dt = \alpha c\rho - \delta$, where δ is the loss referred to unit volume. It is easy to see that even for a constant gain coefficient the quantity varies over the cavity length. Taken as ρ for reasons of simplification is the geometric mean of the values of the radiation density of the cavity mirror. The simplest calculations hence yield the following relationship for the magnitude of the loss:

$$\delta = \frac{\rho c}{L}\left[\frac{1-R}{1+R}\frac{e^{\alpha L}}{2}\right]^{1.2}.$$

The values R = 0.5, L = 10^2 cm, and the transverse discharge dimension l = 2 cm were used in the numerical computations.

The system of kinetic equations in combination with the relationships presented for the electron concentration, gas temperature, and radiation intensity were solved on a BÉSM-6 electronic computer by the Runge–Kutta method. The mean computation time for modification was ~1 min.

Typical curves of the time history for the relative populations ρ_i of the N_2 and CO_2 levels, the relative inversion Δ, the output radiation intensity G, the lasing energy taken off per unit volume of the active medium Q_l, the lasing efficiency η, as well as the temperature T and the relative gas density N/N_0, are represented in Fig. 1. The solid and dashed lines indicate, respectively, the results for models (I) and (II) with a stationary ionization source, while the dot-dashed line corresponds to the results for a pulse ionization source (model I). Curves 1-3 correspond to the relative populations of the levels (010) CO_2, (v = 1)N_2 and (001)CO_2. The peculiarities of the dynamic characteristics presented can be summarized as follows:

1. Both models yield qualitatively similar and quantitatively close results. The rather higher lasing parameters in model (II) are associated with the elevated rate of transmission of the excitation from N_2 to CO_2.

2. The practically linear growth of the relative populations of the levels (v = 1)N_2 and (001) CO_2 after build-up of the generation mode with a stationary ionization source is due to the constancy of the electron excitation rate and the comparatively weak temperature dependence of the rate constants of the reactions $CO_2 + N_2^* \rightleftarrows CO_2^* + N_2$ in the T = (300 + 500)°K range. The relative population of the lower lasing level grows considerably more slowly, which is associated with the rapid transition of excitation in the flexible mode and to the growth in the rate of the V–T-deactivation as the gas temperature rises during generation. The circumstances mentioned determine the almost linear growth of the inversion, to assume the existence of a quasistationary generation mode section despite the temperature rise and the diminution in active medium density. For the conditions presented in Fig. 1, the duration of the quasistationary generation mode τ_s, defined as the time for the radiation intensity to be halved as compared with the steady value at the beginning is around 30 μsec. (The vibrational structure of the initial section of G(t) and other quantities is not of special interest, since it is ordinary consequence of the rapid transition of the system from one quasiequilibrium state to another.) The fact that the lasing efficiency at the time τ_s is close to the maximum value ($\eta_m \simeq 27\%$) and that the energy emitted per unit volume Q_l grows comparatively weakly for $t > \tau_s$ merits attention. It is evident that the time τ_s should correspond to disconnection of the discharge for the practical realization of a lasing system of such a kind.

3. The recombination electron losses for a pulse ionization source result in a sufficiently rapid equalization between the rates of the electron excitation processes and the rates of the vibrational exchange and the V–T-relaxation. This explains the time history of all the appropriate quantities shown in Fig. 1. The results obtained are interesting from the viewpoint of the possibility of using high-voltage pulse generators for ionization of the gas in a nonindependent discharge [2].

The dependence of the main lasing parameters, the efficiency η_s and the mean radiation intensity G_s during the time τ_s, on the magnitude of the electron concentration is presented in Fig. 2 for the quasistationary generation mode. Here the numbers 1, 2, 3 denote the results obtained for the mixture compositions $CO_2 : N_2 : He$ = 15 : 25 : 60; 1 : 1 : 3 and 1 : 1 : 8. The G_s curves are constructed for p_0 = 700 torrs.

The lasing efficiency is practically independent of the pressure for a given mixture composition. The characteristic singularity of the dependence of η_s on the electron density is the presence of a broad maximum. To the left of the maximum, in the low electron concentration domain, the diminution in η_s is associated with the growth in the role of the collision deactivtion process of the upper lasing level. For this reason, the maximum efficiency shifts to the right, while simultaneously diminishing in absolute value (see Fig. 2) as the helium content in the mixture increases.

The diminution in the lasing efficiency as the electron density grows to the right of the maximum is due to the growth in the relative magnitude of the energy losses in excitation of the symmetric and flexible CO_2 modes. It is interesting to note that the existence of this energy contribution channel determines the real value of the limiting efficiency of electrical discharge CO_2 lasers in conformity with the relationship

$$\eta_{max} = \frac{\nu_l}{\nu_3}\left[\sum_i \frac{\nu_i}{\nu_3}\frac{k_i}{(k_3 + k_4)}\right]^{-1},$$

where ν_l is the radiation frequency of the lasing transition, and ν_i and k_i (i=1, 2, 3, 4) are, respectively, the frequencies of the single-quantum transitions and the rate constants of electron excitation of the fundamental modes of the CO_2 and N_2 molecules. Substitution of the values of k_i presented in [3] yields the quantity $\eta_{max} \simeq 30\%$. For sufficiently high electron concentrations ($n/N_C \geqslant 10^{-5}$), recombination losses start to exert a noticeable influence on the magnitude of the lasing efficiency. The character of the dependence of G_S on n (see Fig. 2) is determined practically completely by the factors listed and requires no additional clarification.

For a fixed valve of n/N the quantity G_S depends on the initial gas density according to the law $G_S \sim N_0^2$. Results of computing the quantities G_S/L and τ_S for different values of N_0 and n/N_C are represented as nomograms in Fig. 3. Values of the parameter n/N_{CO_2} and the helium content in the mixture (in parentheses) are indicated for each graph for the relation $CO_2 : N_2 = 1 : 1$. The solid lines correspond to "optimal" laser pumping modes for which the maximum values of the lasing efficiency are realized. Thus, for a $CO_2 : N_2 : He = 1 : 1 : 8$ mixture composition of an initial pressure of 10^3 torr, the duration of the quasistationary generation section will be around 30 μsec under optimal conditions, for an output radiation flux density on the order of $3 \cdot 10^5$ W/cm^2.

Summarizing, let us note that within the framework of a comparatively simple model, the dynamics of CO_2-laser excitation by a combined discharge can be investigated and the parameters of the quasistationary generation mode, whose duration can be regulated by changing the quantity n, can be determined. The most essential singularity of the dependence of lasing system parameters on the magnitude of the electron density for E/N = const is the existence of a domain of values of n/N for which the magnitude of the lasing efficiency is a maximum. The quantitative data obtained about the CO_2-laser characteristics for a broad range of controllable conditions can be used to predict the parameters of lasing systems to be developed with a combined discharge.

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