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The non-self-sustaining gas discharge for exciting continuous-wave gas lasers

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This review devotes its major attention to analyzing the physical processes that govern the stability of uniform burning of the volume gas discharges that are used to excite high-pressure CO_2 lasers. An analysis is given of the theoretical and experimental studies that have shown a substantial increase in the time of uniform burning of a non-self-sustaining gas discharge as the electric power density is decreased. Specificially, this has made it possible to convert the discharge in practice into a steady-state burning regime at elevated gas pressure. The optical characteristics of gas mixtures based on carbon dioxide that are established upon excitation of the medium by a steady-state non-self-sustaining discharge are examined.

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CONTENTS

1. INTRODUCTION

The interest in CO_2 lasers that operate at elevated pressures of the working medium (of the order of a hundred torr and higher), and which are excited by a non-self-sustaining gas discharge, is explained by the possibility of attaining high energy densities and efficiencies of laser action in these systems.

The gas discharge as used for exciting CO_2 lasers is a glow discharge whose plasma is very weakly ionized: the fraction of charged particles with respect to the concentration of atoms and molecules is of the order of 10^{-6}–10^{-8}. The temperature of the electrons in the gas-discharge plasma as established by the action of the electric field is about 1 eV, while the temperature of the gas is close to room temperature. The thermodynamic non-equilibrium of the plasma, which is specifically what creates the inverted population of the vibrational levels of the CO_2 molecules, is determined by the greater heat capacity of the gas than that of the charged particles. Yet the heating of the neutral particles that occurs during the burning of the discharge limits the duration of laser action in pulsed lasers. Heat is usually removed from the gas in continuous-wave lasers by pumping the gas through the discharge volume.

In a self-sustaining discharge, the electric field that is established in the discharge, and correspondingly,

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the value of the electron temperature, is governed by the ionization properties of the medium. It is maintained at a level that allows the electrons of the plasma to effect the ionization of the atoms and molecules required for burning of the discharge. As compared with these discharges, the non-self-sustaining gas discharge that is used for exciting CO_2 lasers has two fundamental advantages that are interrelated to some extent.

First, the use of an external ionizer to create the gas-discharge plasma permits one to vary the electric-field intensity of the gas discharge over a wide range without restriction to the quite fixed values of the working field intensity that are inherent in self-sustaining discharges. One can thereby keep a gas discharge burning in the range of electric fields that create an electron distribution function in the plasma that is most suitable for populating the working laser levels. This situation, which was first noted in Ref. 1, is especially important in exciting gases in which the energy of the upper laser level is substantially lower than the ionization potential. This is because the working field intensity of the self-sustaining discharge that is required to maintain the appropriate rate of ionization of the gas can prove to be too high, and to exceed the value at which the excitation of the low-energy laser levels is optimal.

This is just the situation that occurs in lasers based on carbon dioxide: the energy of the upper laser level is 0.3 eV, while the ionization potential of the atoms and molecules that are usually present in the gas mixture is about 15 eV. Hence the use of a non-self-sustaining discharge to excite CO_2 lasers makes possible the most efficient conversion of electrical energy into coherent-radiation energy.

A second advantage of the non-self-sustaining gas discharges that are used to excite CO_2 lasers is their stability and uniformity of burning as compared with that of self-sustaining discharges. This difference in the type of course of the gas discharges stems from the fact that the electron concentration in the plasma created by an external ionizer is determined by the power of this ionizer. Therefore fluctuations in the electron temperature do not lead to strong fluctuations in the electron concentration, and accordingly to redistribution of the gas-discharge current.

This fact of enhanced stability and uniformity of burning of the gas discharge when the gas-discharge plasma is not created by its own electric field but by an external ionizer was first noted in Ref. 2, which was concerned with studying the negative-glow region of a self-sustaining glow discharge. In essence, this is a region where a non-self-sustaining burning regime is always maintained that is caused by ionization of the gas by fast electrons that leave the cathode layer, and which are accelerated by the voltage fall near the cathode.

A practically important manifestation of these physical properties of non-self-sustaining discharges was the possibility of increasing the gas pressure in CO_2 lasers excited by a non-self-sustaining discharge. Here the conditions cited above for maintaining non-self-sustaining discharges permit one to optimize the excitation of the laser levels of the CO_2 molecules by independently varying the applied electric field and the gas composition over wide ranges. This potentiality has been demonstrated by an entire series of investigators. The use of powerful ionization sources (high-current electron accelerators,[3-9] a pulsed reactor,[10] sources of intense ultraviolet radiation[11]) has allowed them to build powerful pulsed lasers based on carbon dioxide operating at a gas pressure of the order of atmospheric or higher. The duration of uniform burning of the non-self-sustaining gas discharge in these lasers, and correspondingly, the duration of laser action, did not exceed several microseconds. This stemmed from the high conductivity of the medium and the consequent high power input, which rather rapidly overheated the medium. In this sense, these lasers resemble lasers having a transverse self-sustaining discharge (TEA-lasers), in which uniform development of the discharge in gases at elevated pressures is initiated either by choosing a special design of the electrodes (sectioned electrodes),[12-14] or by applying to the interelectrode gap very fast large overvoltages that give rise to multi-streamer breakdown[15-17]; or by preionization of the medium, which is carried out in highly varied ways.[18-27] An analysis of the studies on TEA-lasers and pulsed CO_2 lasers excited with powerful ionizers is found in the reviews[28,29].

Yet there is an entire set of reasons why the use of ionizers of such high power does not allow one to proceed to designing continuous-wave lasers having an elevated gas pressure. First, the use of these ionizers, which overheat the medium in a time of ~1 μsec, requires pumping of the gas through the working volume of the laser at very high speeds. Thus, even with a linear dimension of the laser of ~1 cm along which the gas is to be pumped, one must have a velocity of gas flow of $\sim 10^6$ cm/sec. It is extremely hard to achieve technically a flow of gas at velocities exceeding the speed of sound by factors of tens. Second, a current technically insurmountable difficulty in using such powerful ionizers for designing continuous-wave gas lasers is that of attaining steady-state, rather than pulsed, operation of these ionizers. For example, conversion of high-current electron accelerators to a continuous regime of operation is hindered by overheating and by breakdown of the thin films that separate the vacuum space of the accelerator from the volume of the discharge chamber of the laser.[1)]

One can substantially facilitate the solution of these problems that arise in designing continuous-wave gas lasers having an elevated pressure of the working medium by decreasing the power level of the external ionizer, and correspondingly decreasing the power density input into the non-self-sustaining gas discharge. Naturally, the feasibility of this way of designing continuous-wave gas lasers is determined to a considerable extent by the characteristics of non-self-sustaining gas

[1)]A detailed analysis of the problems that face the technology of designing CO_2 lasers excited by electron accelerators is given in the review, Ref. 30.

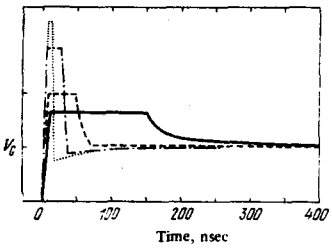

FIG. 1. Voltage oscillograms obtained in Ref. 25a for various values of the initial voltage applied to the gas-discharge gap.

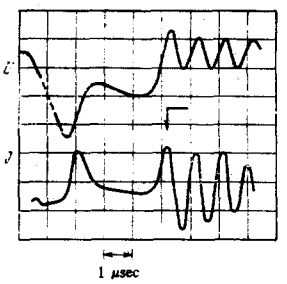

FIG. 2. Voltage ($U = 20$ kV/division) and current ($I = 1$ kA/division) oscillograms obtained in Ref. 42 for a discharge in a CO_2–N_2–He gas mixture at atmospheric pressure. The arrow indicates the instant of conversion of the uniform glow discharge into a localized arc.

discharges in dense gases that are manifested when the power of the ionizing radiation is substantially decreased. These problems have been studied intensively in recent time, beginning with Ref. 31, in which uniform burning of a steady-state non-self-sustaining discharge having electronic conduction in a subsonic gas flow at atmospheric pressure was first shown possible.

This review will devote major attention to analyzing the physical processes in the plasma of a non-self-sustaining discharge, which in its characteristics can be easily used for designing powerful continuous-wave gas lasers. Also, to allow comparison of the physical processes that determine the stability of burning of a glow discharge in the self-sustaining and non-self-sustaining regimes, this review will briefly analyze the fundamental results of studies on ionization instabilities that develop in the self-sustaining glow discharge, and which rapidly convert these discharges into a contracted arc.

2. THE UNIFORM SELF-SUSTAINING GLOW DISCHARGE IN GASES AT ELEVATED PRESSURE

Uniform burning of a glow discharge at high pressures (of the order of hundreds of torr and higher) is unstable. In the course of a certain time, a filament structure is formed in the discharge, and the discharge contracts and converts into an arc. Here the very existence of the initial phase of uniform burning of the glow discharge at high pressure requires special conditions for initiating the discharge. One must preionize the gas in order to start a uniform high-pressure discharge.[2)] When an electric field of intensity close to the breakdown value is applied to the preionized gas, the discharge burns uniformly for some time, while showing features intrinsic to diffuse glow discharges[37–40] (a Faraday dark space, a bright negative glow near the cathode, and a uniform positive column). The minimum initial electron density n_0 needed for uniform burning is given by the relationship[14,41]

$$n_0^{-1/3} \leqslant \left(\frac{\langle v\rangle \lambda z_{cr}}{v_e}\right)^{1/2}, \qquad (1)$$

where λ is the free flight path of the electrons; v_e and $\langle v\rangle$ are their drift and thermal velocities; and z_{cr} is the distance that the avalanche travels in the direction of the electric field in the time that it takes for the internal field of the avalanche to reach the applied field value.[32] In deriving Eq. (1), the criterion for uniform breakdown was taken to be the overlap of all the developing avalanches as they approach the instant of converting into streamers. This is because here the perturbation of the field by the growing space charge will be the most uniform, and it should not intensify the development of breakdown in the form of a single streamer. For a pressure of the order of atmospheric and with the appropriate values of the breakdown electric field intensity in the gas mixtures of CO_2 lasers, the estimate of (1) gives as the minimum value of the initial electron density $n_0 = 10^4$–10^5 cm^{-3}.

Use was made of the "double discharge" scheme proposed in Ref. 18 to preionize the gas, as well as irradiation of the cathode[19–21] and the discharge gap[22–27] with ultraviolet. Figures 1–3 illustrate the dynamics of development of this discharge. Figure 1[25a] shows oscillograms of the gas-discharge voltage in CO_2-N_2-He gas mixtures at atmospheric pressure. These oscillograms correspond to the burning of the gas discharge up to the instant of formation of an arc. Figure 2 shows the characteristic shape of the current and voltage oscillograms, which indicate the establishment of uniform burning and subsequent conversion to an arc.[42] Figure 3[16] shows an oscillogram of the current and photographs of continuous and time-lapse pictures of the emission from the discharge in He–CO_2 gas mixtures at a atmospheric pressure. We see from Figs. 1–3

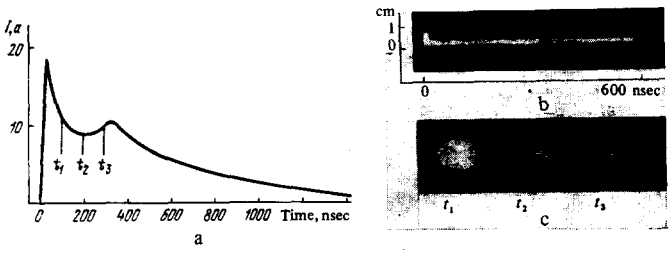

FIG. 3. a) Oscillogram of the discharge current in a He/CO_2 mixture at atmospheric pressure as obtained in Ref. 16; b) continuous slit photoscan of the cross-section of the discharge; c) time-lapse photograph of the discharge. The cathode lies on the right-hand side of each frame.

[2)]Other ways of starting a uniform high-pressure discharge involve an appropriate choice of the size of the interelectrode gap in which a Townsend mechanism of breakdown occurs,[32–40] or applying high overvoltages to the discharge gap,[15–17] or sectioning the electrodes.[12–14]

that both the electrical and the plasma characteristics of the discharge in dense gases remain unchanged for some time after formation.

The fundamental factor that restricts the time of existence of the high-pressure homogeneous pulsed discharge is overheat-ionization instability.[43-47] It converts the discharge into an arc that is sharply nonuniform over the volume. The mechanism of this phenomenon involves the sharp dependence of the ionization constant on the ratio E/N of the electric field intensity in the discharge to the gas density. Since diffusion of electrons plays practically no role under high-pressure-discharge conditions, the local density of electrons is proportional to the ionization constant, and hence so is the intensity of heat release. Hence small fluctuations in the gas density arising from the described mechanism of heat release are sharply amplified. This redistributes the current density over the volume of the discharge.

Let us estimate the characteristic value of the increment of the stated instability by starting with the following treatment of the development of filamentous fluctuation in the gas density parallel to the electric field.[44] Let the characteristic dimension of a random inhomogeneity l satisfy the conditions:

$$\lambda_t \ll l \ll c_s \tau_{ch}, \tag{2}$$

Here c_s is the speed of sound, λ_t is the characteristic dimension of the thermal conductivity of the gas, and τ_{ch} is the characteristic time of development of instability.

The left-hand inequality in (2) allows one to neglect the heat conduction transverse to the developing current filament. Owing to the right-hand inequality (the velocity of movement of the gas is small in comparison with the velocity of sound), we can assume the gas pressure in the developing current filament to be constant. Therefore, we can restrict the treatment in analyzing the system of hydrodynamic equations to the equations of continuity and energy balance:

$$\frac{\partial N}{\partial t} + N \operatorname{div} \mathbf{v} = 0, \tag{3}$$

$$\frac{\gamma}{\gamma - 1} P \operatorname{div} \mathbf{v} = e\mu_e n_e E^2; \tag{4}$$

Here P and $\mathbf{v}$ are respectively the pressure and velocity of movement of the gas, γ is the exponent of the adiabatic curve, n_e and μ_e are the concentration and the mobility of the electrons, and e is the electron charge.

As we have noted, diffusion of electrons is not substantial under the studied conditions, and hence the local electron density is proportional to the ionization constant:

$$n_e = f(E/N) \exp\left(-\frac{BN}{E}\right), \tag{5}$$

Here B is an empirical coefficient and f is some power function that is obtained with account taken of the approximation of the ionization coefficient by $\alpha \sim \exp(-BN/E)$.[48]

Equations (3)–(5) lead to an equation that describes the local decrease in the density of the gas caused by its Joule heating upon collision of the electrons with the neutral particles:

$$\frac{dN}{dt} = -\frac{\gamma - 1}{\gamma} \frac{Nf \exp(-BN/E) e\mu_e E^2}{P}. \tag{6}$$

We can write for this equation a qualitative solution that accounts only for the exponential dependence, in the form

$$N = \frac{E}{B} \ln\left[\exp\left(\frac{BN_0}{E}\right)\left(1 - \frac{BN_0}{E} \frac{\sigma_0 E^2}{P} \frac{\gamma - 1}{\gamma} t\right)\right]; \tag{7}$$

here σ_0 and N_0 are the conductivity and the density of the gas at the initial instant of time. Here, as we see from (5), the concentration of electrons varies according to the law

$$n_e = \frac{n_{e0}}{1 - (t/\tau_{ch})}. \tag{8}$$

We see from the obtained solution that overheat-ionization instability, which substantially decreases the local gas density and sharply increases the electron density, develops explosively with a characteristic time

$$\tau_{ch} \simeq \frac{E}{BN_0} \frac{\gamma}{\gamma - 1} \frac{P}{\sigma_0 E^2} \equiv \frac{1}{c}. \tag{9}$$

We see that the given estimate of τ_{ch} does not depend on l, the characteristic dimension of the random inhomogeneity. Naturally this holds for short-wavelength perturbations as long as the right-hand inequality of (2) is satisfied. Yet, with increasing l a gas-pressure gradient arises transverse to the developing filamentary perturbation. Then one can estimate τ_{ch} by linearizing the initial system of equations with respect to relatively small perturbations growing on a uniform non-steady-state background.[47] The initial non-steady-state equations of continuity, motion, and energy of the gas have the following form:

$$\frac{\partial \rho}{\partial t} + \frac{\partial (\rho v)}{\partial x} = 0, \tag{10}$$

$$\frac{\partial v}{\partial t} + v \frac{\partial v}{\partial x} = -\frac{1}{\rho} \frac{\partial P}{\partial x}, \tag{11}$$

$$\frac{\gamma}{\gamma - 1} \rho\left[\frac{\partial}{\partial t}\left(\frac{P}{\rho}\right) + v \frac{\partial}{\partial x}\left(\frac{P}{\rho}\right)\right] - \frac{\partial P}{\partial t} - v \frac{\partial P}{\partial x} = Q \equiv \sigma E^2; \tag{12}$$

Here P, ρ, and v are respectively the pressure, mass density, and velocity of the gas; and x is the direction normal to the direction of the gas-discharge current. The conductivity (σ) of the weakly-ionized plasma is mainly determined by the electron concentration in the positive column, and it can be represented in the form:

$$\sigma = e\mu_e n_e = \varphi(E) \rho^{-a}. \tag{13}$$

Here $\varphi(E)$ is some function of the electric field intensity; $a \gg 1$ is some mean exponent that approximates the relationship of the ionization coefficient to the parameter E/ρ.

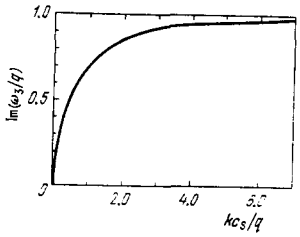

FIG. 4. Relationship of the increment of overheat-ionization instability to the wave number of a developing perturbation.[47]

We can expand the variables P, ρ, and v that enter into Eqs. (10)–(12) with respect to a developing spatially uniform state as follows:

$$P(x, t) = P_0(t) + P_1(t) e^{ikx},$$

$$\rho(x, t) = \rho_0 + \rho_1(t) e^{ikx},$$

$$v(x, t) = v_1(t) e^{ikx},$$

here the subscript 0 denotes the unperturbed non-steady-state values that are established in the developing discharge; the subscript 1 denotes the perturbations whose development particularly implies instability of the initial spatially-uniform state; and k is the spatial wavenumber of the studied perturbations.

The system (10)–(12) as linearized with respect to the smallness of the perturbations has the form

$$\frac{\partial \rho_1}{\partial t} + ik\rho_0 v_1 = 0, \tag{10a}$$

$$\rho_0 \frac{\partial v_1}{\partial t} = -ikP_1. \tag{11a}$$

$$\frac{1}{\gamma-1}\frac{\partial P_1}{\partial t} - \frac{\gamma}{\gamma-1}\frac{P_0}{\rho_0}\frac{\partial \rho_1}{\partial t} = -Q_0 a \frac{\rho_1}{\rho_0}. \tag{12a}$$

Upon eliminating all the variables but ρ_1 from these equations, we get

$$\frac{d^3\rho_1}{dt^3} + k^2 c_s^2 \frac{d\rho_1}{dt} - \frac{a(\gamma-1)k^2 Q_0}{\rho_0}\rho_1 = 0. \tag{14}$$

Here $c_s = \sqrt{\gamma P_0/\rho_0}$ is the speed of sound. Now if we assume that $\rho_1(t) \propto \exp(-i\int \omega dt)$ and neglect the higher derivatives of ω, we can write the dispersion relationship

$$i\omega(\omega^2 - k^2c_s^2) - 3\omega\frac{d\omega}{dt} - \frac{a(\gamma-1)}{\gamma}\frac{k^2c_s^2 Q_0}{P_0} = 0. \tag{15}$$

If $|d\omega/dt| \ll \omega^2$, then the roots of Eq. (15) are determined by the expressions

$$\left.\begin{aligned} \omega_1 &= (S_+ - S_-)\frac{\sqrt{3}}{2} - \frac{i}{2}(S_+ + S_-), \\ \omega_2 &= -(S_+ - S_-)\frac{\sqrt{3}}{2} - \frac{i}{2}(S_+ + S_-), \\ \omega_3 &= i(S_+ + S_-), \end{aligned}\right\} \tag{16}$$

where

$$S_\pm = \left[\frac{1}{2}k^2c_s^2 q \pm \left(\frac{1}{27}k^6c_s^6 + \frac{1}{4}k^4c_s^4 q^2\right)^{1/2}\right]^{1/3}, \qquad q = \frac{a(\gamma-1)Q_0}{\gamma P_0}.$$

We see from the expressions (16) that the two first (acoustic) perturbation modes decay in time, while the third mode grows. This corresponds to the growth of overheat-ionization instability.

The performed treatment, which took account of the perturbation of the gas pressure, gives the relationship of the increment of overheat-ionization instability to the wavelength of the perturbation. Figure 4 shows $\mathrm{Im}(\omega_3/q)$ as a function of kc_s/q.[47] We see that instability develops more slowly with increasing wavelength of the perturbation. As we should expect, the increment is saturated for short-wavelength perturbations at the level

$$\mathrm{Im}\,\omega_3 = q \equiv \frac{a(\gamma-1)}{\gamma}\frac{Q_0}{P_0}. \tag{9a}$$

This expression for $\tau_{ch} = 1/\omega_3$, which holds for $kc_s/q \gg 1$, naturally coincides with Eq. (9), which was derived for short-wavelength perturbations. The nature of the overheat-ionization instability in various concrete situations has been analyzed in detail in Refs. 43, 45, 46.

We assumed above in analyzing the development of overheat-ionization instability that the electrons transfer their energy rather quickly to neutral particles. Therefore the Joule energy input was substituted into the energy balance equation as the energy source: $Q = \sigma E^2$. In a discharge in atomic gases, we can assume that all the energy acquired by the electrons in the electric field goes into heating the gas if the time τ_{e0} that it takes for an electron to impart its energy to atoms via elastic electron-atom collisions is small in comparison with τ_{ch}. Upon using Eq. (9), we can write the condition for $\tau_{e0} \ll \tau_{ch}$:

$$\frac{M}{m}\frac{1}{\nu_e} \ll \frac{E}{BN_0}\frac{P}{\sigma_0 E^2}, \tag{17}$$

Here M and m are respectively the mass of the atom and of the electron, and ν_e is the frequency of the electron-atom collisions.

The pattern of development of overheat-ionization instability in a discharge in a molecular gas is somewhat complicated by the existence of the vibrational degree of freedom of the molecules. The energy of the electrical source is initially converted into vibrational energy of the molecules, and then, owing to the slower process of VT relaxation, it is released in the form of heat. Hence the condition analogous to (17) is given by the expression

$$\tau_{VT} \ll \frac{E}{BN_0}\frac{P}{\sigma_0 E^2}, \tag{18}$$

where τ_{VT} is the vibrational-translational relaxation time.

In the gas mixtures that serve as the working media of CO_2 lasers, τ_{VT} often has a value that violates the condition (18). Hence, in analyzing the development of overheat-ionization instability in this case, we must include in the treatment the kinetics of population of the vibrational levels of the molecules whose relaxation is specifically the source of the heating of the gas.[46] Then, upon considering that the bulk of the energy of the electrons is converted into the vibrational energy of molecules, while the vibrational-translational relaxa-

tion time is large in comparison with the characteristic time of the process, we can write the equations of energy balance for the excited molecules and for the gas temperature in the developing discharge:

$$\rho\frac{\partial\varepsilon}{\partial t}+\rho v\frac{\partial\varepsilon}{\partial x}=\sigma E^2\equiv Q, \tag{19}$$

$$\frac{\gamma}{\gamma-1}\rho\left[\frac{\partial}{\partial t}\left(\frac{P}{\rho}\right)+v\frac{\partial}{\partial x}\left(\frac{P}{\rho}\right)\right]-\frac{\partial P}{\partial t}-v\frac{\partial P}{\partial x}=\frac{\varepsilon\rho}{\tau_{VT}(T)}\equiv W. \tag{20}$$

Here ε is the specific vibrational-energy density of the molecules. Putting $\varepsilon(x,t)=\varepsilon_0(t)+\varepsilon_1(t)e^{ikx}$, we linearize Eqs. (19) and (20) in terms of the smallness of the perturbations that arise on the non-steady-state spatially uniform background:

$$\rho_0\frac{d\varepsilon_1}{dt}=-\frac{aQ_0}{\rho_0}\rho_1, \tag{19a}$$

$$\frac{1}{\gamma-1}\frac{\partial P_1}{\partial t}-\frac{\gamma}{\gamma-1}\frac{P_0}{\rho_0}\frac{\partial\rho_1}{\partial t}=\frac{\varepsilon_1\rho_0}{\tau_{VT0}}-\frac{\varepsilon_0 M}{\tau_{VT0}^2}\left(\frac{d\tau_{VT}}{dt}\right)_0 P_1+\frac{\varepsilon_0 P_0 M}{\rho_0\tau_{VT0}^2}\left(\frac{d\tau_{VT}}{dt}\right)_0\rho_1. \tag{20a}$$

Here M is the mass of the heavy particles. We assumed in deriving these equations that the conductivity σ of the plasma is represented by (13) with $a\gg1$. We also assumed that $-d\ln\tau_{VT}/d\ln T\equiv b\gg1$, since $\tau_{VT}(T)$ is usually a strongly declining function.[49]

We can use the derived equations (19a) and(20a) as well as (10a) and (11a) to get a dispersion equation in the studied case that is analgous to Eq. (14). Yet we shall restrict ourselves here to the following qualitative treatment that permits estimating the minimum characteristic time of development of overheat-ionization instability in the studied case.

Naturally, as in the previously analyzed case (for $\tau_{VT}\ll\tau_{ch}$), perturbations have the minimum development time when their wavelengths satisfy the inequality $kc_s\tau_{ch}\gg1$. Upon taking account of the fact that $P_1\simeq0$ in such perturbations, and deriving from (19a) the estimating relationship $\varepsilon_1=-(aQ_0\tau_{ch}/\rho_0^2)\rho_1$, we transform the right-hand side of Eq. (20a) as follows:

$$W_1=-\frac{(a+b)Q_0\tau_{ch}}{\tau_{VT0}}\frac{\rho_1}{\rho_0}. \tag{21}$$

Upon comparing this expression with the right-hand side of Eq. (12a), we can by analogy with (9a) derive a relationship that determines the characteristic development time of instability for short-wavelength perturbations

$$\tau_{ch}\approx\frac{P_0\tau_{VT0}}{(a+b)Q_0\tau_{ch}}.$$

Finally, this gives rise to an expression that determines the characteristic development time of overheat-ionization instability in the studied case when $\tau_{VT0}\gg\tau_{ch}$:

$$\tau_{ch}\approx\sqrt{\frac{P_0\tau_{VT0}}{(a+b)Q_0}}. \tag{22}$$

Thus, in a discharge in molecular gases, in which the bulk of the energy of the electrons is converted into vibrational energy of the molecules, the time τ_{ch} is observed to decline with increasing Joule energy-input power, as determined by Eq. (9), until it becomes shorter than the vibrational-translational relaxation time of the molecules, which then governs the heating of the gas. When this Joule energy-input power is reached, the characteristic time of development of overheat-ionization instability in molecular gases is found from Eq. (22). Interestingly, in this case the development time of instability declines with increasing Joule energy-input power $\sim1/\sqrt{Q_0}$, in contrast to the linear inversely proportional relationship that one gets in the approximation of instantaneous energy transfer from the electrons to the temperature of the gas. We also see from Eq. (22) that the more strongly the relationship $\tau_{VT}(T)$ declines, the more rapidly instability develops.

Another type of ionization instability can develop in a self-sustaining discharge that excites CO_2 lasers; in contrast to overheat-ionization instability, this type does not involve heating the gas. The mechanism of this instability, which has been studied in Refs. 14, 50, 51, involves the fact that the degree of excitation of the vibrational levels of the molecules increases with increasing energy of the electrons. In turn this diminishes the energy losses of the electrons, and thus increases their energy. Having a greater energy, the electrons cause stronger ionization. Consequently a fluctuational current instability develops. Just as for overheat-ionization instability, the development of this instability is explosive in nature.[51] Yet its time of development is naturally determined by the characteristic time for excitation of the vibrational energy of the molecules:

$$\tau_k\approx\frac{\tilde{a}I}{aQ_0}, \tag{23}$$

where I is the energy of a vibrational quantum; and the coefficient $\tilde{a}=d\ln n_e/d\ln T_e$ characterizes the strong dependence of the concentration of the electrons on their temperature.

Upon comparing Eqs. (23) and (22), we can say that the ionization instability of the discharge in molecular gases arises from saturation of the channel of excitation of the vibrational levels of the molecules by the electrons if

$$\frac{\tau_{ch}}{\tau_k}\approx\frac{\tilde{a}\sqrt{Q_0}}{\sqrt{a-b}\,\tilde{a}I}\sqrt{P_0\tau_{VT0}}>1. \tag{24}$$

Since $Q_0\sim n_e$, the inequality (24) determines the minimum electron concentration only if the studied instability develops in a pure form when this concentration is exceeded. However, overheat-ionization instability will develop sooner if the electron concentration in the discharge is lower.

For conditions typical in self-sustaining discharges for exciting CO_2 lasers ($Q_0/P\gtrsim10^5$ W/cm^3 · atm, $n_e>10^{14}$ cm^{-3} at $P\sim1$ atm), Eqs. (9) and (23) give values of τ_{ch} and $\tau_k\lesssim10^{-6}$ sec. This value is characteristic of the experimentally observed time of conversion of uniform self-sustaining discharges into arcs in gases at elevated pressures.

In addition to the rather general mechanisms of in-

stability that we have discussed, special conditions can in principle be realized in the plasma of a self-sustaining gas discharge that favor development of a number of other instabilities that convert a uniform glow discharge into an arc.[37b,38,39,46,52-54] The characteristic times of development of these instabilities in a gas at atmospheric pressure are also $\sim 10^{-6}$ sec.

Thus the analysis performed in this section shows that the characteristic time of conversion of uniform self-sustaining discharges into arcs in gases at elevated pressure is small ($\lesssim 10^{-6}$ sec). It is determined, first, by the strong exponential relationship of the concentration of electrons to their temperature, and second, by the high power of the Joule energy Q_0 that is released in the plasma at characteristic burning voltages. This is just why shifting to a non-self-sustaining discharge, with the possibility of varying the electrical power released in this discharge over a broad range, creates the prerequisites for substantially increasing the time of stable, homogeneous burning of the discharge in gases at elevated pressure.

3. BURNING AND STABILITY OF THE NON-SELF-SUSTAINING GAS GLOW DISCHARGE

A. Physical processes in the non-self-sustaining gas discharge

The possibility of reducing the power of the external ionizer to values that will permit converting this ionizer and the non-self-sustaining discharge in a subsonic flow of a dense gas into a continuous mode of operation was first studied in Ref. 31. Following this study, let us examine the conditions at the minimum possible power of the external ionizer for which the discharge is still non-self-sustaining, while the electric current arises from electron transport.[3)]

The following conditions must be satisfied for maintaining a non-self-sustaining gas discharge in which the current arises from electron transport. First, the size of the cathode fall that brings about the emission of electrons required to maintain the current must be small in comparison with the voltage applied to the discharge gap. Second, the rate of production of electrons in the region of the gas discharge away from the cathode under the action of the external ionizer must exceed the rate of the intrinsic electron multiplication under the action of the applied electric field. These conditions ensure that a discharge current will flow in electric fields below the breakdown value. This eliminates the prerequisites for nonuniform burning that characterize the initial stage of discharges at high pressures, and it also eliminates the factors that lead to development of ionization instabilities that were discussed in the last section.

First let us find what are the minimum possible rates of production of electrons by the external ionizer, and hence also, the minimum possible energy-input powers at which the discharge will remain non-self-sustaining. The condition for a non-self-sustaining discharge is that the ionization in the gas volume is mainly due to the external ionizer rather than to the intrinsic electron multiplication in the electric field of the gas discharge. It is given by the expression

$$S \gg \alpha n_e v_e; \tag{25}$$

Here S is the rate of production of electrons per unit volume of gas by the external ionizer; and α is the number of electron-ion pairs produced per unit distance by the electrons of the plasma themselves.

The ionization coefficient $\alpha/P(E/P)$ was calculated in Refs. 25a, 53, 56 for a series of CO_2–N_2–He gas mixtures. These studies indicate that α varies by 5–7 orders of magnitude in the range of values of $E/P = 3$–10 V/cm·Torr in which the distribution function of the electrons is optimal for exciting the vibrational levels of the nitrogen molecules and the upper laser level of the CO_2 molecules.[57] Thus we can conclude that the inequality (25) allows a quite substantial decrease in the power of the external ionizer with a small decrease in E/P. For example, one can easily make estimates[31] that show that condition (25) is satisfied in a discharge in molecular nitrogen at atmospheric pressure with an external ionizer that yields a Joule energy-input power at the level of 10^3 W/cm^3. This value smaller by a factor of $\sim 10^2$ than the corresponding values that were realized in Refs. 3–11. Therefore the use of ionizers at this power level should substantially increase in duration of stable, uniform burning of the non-self-sustaining discharge.

Further decrease in the power level of the external ionizer proves ineffectual owing to the insufficient rate of exciting CO_2 molecules to the upper laser level. The problems involved in creating an inverted population of CO_2 molecules in the working medium of the CO_2 laser will be treated in further detail in Chap. 4.

In order to estimate the size of the cathode drop at relatively small values of the Joule energy-input power (at the level of $\lesssim 10^3$ W/cm^3·atm), i.e., in order to determine the region of existence of the non-self-sustaining discharge in which the current arises from the mobility of electrons, the following system of equations was solved in Ref. 31 that describes the steady-state processes of transport of electrons and ions in molecular nitrogen[4)]:

$$\frac{d(n_e v_e)}{dx} = -\frac{d(n_i v_i)}{dx} = S + \alpha n_e v_e - \beta n_e n_i, \qquad \frac{dE}{dx} = 4\pi e (n_i - n_e), \tag{26}$$

[3)] As in well known, non-self-sustaining discharges having ionic conduction caused by charge separation in the plasma and by strong shielding of the applied electric field (the ionization-chamber regime[55]) can arise at any values of the external-ionizer power and at voltages below the breakdown value. However, these discharges are completely unsuitable for exciting CO_2 lasers, since almost all the electrical energy in them goes into heating the gas, rather than into exciting the vibrational levels of the molecules.

[4)] The cathode potential drop in a non-self-containing discharge has also been calculated in Refs. 4b, 48, and 59.

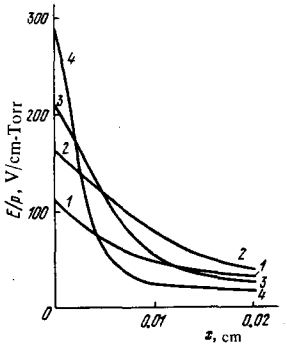

FIG. 5. Electric-field distribution for a discharge in nitrogen at atmospheric pressure (cathode: $x = 0$).[31]

1—$S = 10^{16}$,	$E_0/P = 4$,	$\gamma_i = 10^{-1}$;
2—$S = 10^{16}$,	$E_0/P = 4$,	$\gamma_i = 10^{-3}$;
3—$S = 10^{16}$,	$E_0/P = 10$,	$\gamma_i = 10^{-3}$;
4—$S = 2 \cdot 10^{17}$,	$E_0/P = 10$,	$\gamma_i = 10^{-2}$;

S is in cm^{-3} sec^{-1}, and E_0/P in V/cm·Torr.

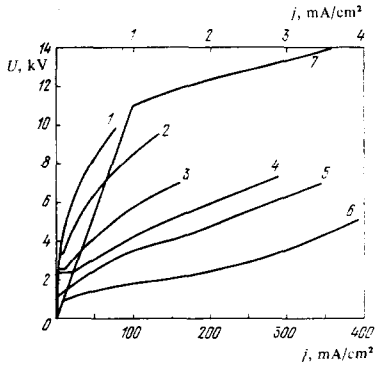

FIG. 7. Voltage-current characteristic of a non-self-sustaining discharge at a pressure of 1 atm.[31] 1—N_2: $CO_2 = 3:2$; 2—N_2: $CO_2 = 4:1$, 2—N_2: $CO_2 = 10:1$; 4—N_2: 5—N_2, oxidized cathode (beam current = 10 μA for curves 1–5); 6—N_2, oxidized cathode, beam current = 40 μA; 7—air (the upper scale of current values refers to curve 7), $v = 62$ m/s.

where x is the direction from the cathode to the anode, v_i is the drift velocity of the ions, and β is the electron-ion recombination coefficient. The assigned boundary conditions were the electron-emission current at the cathode and the electric field in the region of the discharge away from the cathode:

$$n_e(0)\, v_e(0) = -\gamma_i n_i(0)\, v_i(0), \quad E(x \to \infty) \to E_0, \qquad (27)$$

and γ_i is the second Townsend coefficient, which was assumed independent of the energy of the ions. The values of all the coefficients were taken from Ref. 60.

Figure 5 gives the numerical solutions of the Eqs. (26) with the boundary conditions (27) for various values of E_0, γ_i, and S (and hence also for various values of n_e in the main region of the gas discharge). By using the obtained solutions, we can determine the value of the cathode potential fall U_c and the dimension l_c of the cathode layer. For example, at $P = 1$ atm, $E_0/P = 10$ V/cm·Torr, $\gamma_i = 10^{-2}$, and $N_e = 2 \times 10^{11}$ and 10^{12} cm^{-3}, we have respectively: $U_c = 1300$ V, $l_c = 10^{-2}$ cm; and $U_c = 750$ V, $l_c = 5 \times 10^{-3}$ cm. We note that when $n_e = 2 \times 10^{11}$ cm^{-3} and $E_0/P = 10$ V/cm·Torr, the Joule energy-input power is $Q_0 = en_e v_e E \simeq 10^3$ W/cm^3.

The estimates made show that one can maintain a non-self-sustaining discharge in a gas at atmospheric pressure at an interelectrode spacing of the order of several centimeters, and hence, at values of the applied electric field of the order of several tens of kilovolts, with a specific energy-input power of about 1 kW/cm^3. Here the cathode fall constitutes a small fraction of the total applied potential. Hence the size of the discharge current is determined by the electronic conductivity created by the external ionizer.

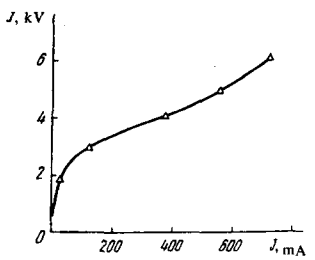

FIG. 6. Voltage-current characteristic of a non-self-sustaining discharge in nitrogen.[61]

The relatively small size of the cathode fall in non-self-sustaining discharges at the studied level of electric power released in the gas-discharge volume has been experimentally confirmed in Refs. 31, 61, and 62a. Figure 6 shows the voltage-current characteristic obtained in Ref. 61 of a non-self-sustaining discharge in molecular nitrogen at the pressure $P = 500$ Torr. The authors used as the external ionizer an ultraviolet source that created an electron concentration at the level of $\sim 10^{11}$ cm^{-3} in the discharge volume. Efficient volume photoionization was achieved here by adding to the nitrogen cesium vapor, which has a low ionization potential (the pressure of cesium vapor amounted to $\sim 10^{-3}$ Torr).[5)]

Figure 7 depicts the family of voltage-current characteristics obtained in Ref. 31 from a non-self-sustaining discharge in N_2–CO_2 gas mixtures at atmospheric pressure. The external ionizer used in this study was a stationary beam of high-energy electrons. The electron energy was ~100 keV, and the current density of the electrons in the beam was ~ 10 μA/cm^2. Estimates show that in this case the concentration of electrons in the gas-discharge plasma is $\sim 10^{11}$–10^{12} cm^{-3}. The observed decline in the electron concentration as the partial content of CO_2 was raised (or when the discharge was conducted in air), and accordingly that in the conductivity of the plasma, is explained by the attachment of electrons to CO_2 or O_2 molecules.

We see from Figs. 6 and 7 that all the voltage-current characteristics show a marked change in slope in

[5)]The possible use of ultraviolet to maintain a non-self-sustaining discharge in gases at elevated pressure with added alkali-metal vapor was first studied in Ref. 63.

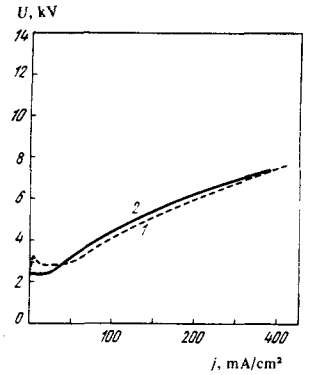

FIG. 8. Calculated (1) and experimental (2) voltage-current characteristics of a non-self-sustaining discharge in nitrogen at atmospheric pressure.[31]

the low-current region. This fact is explained by the idea that the electric field in the plasma is almost fully shielded (concentrated in the cathode layer) at low voltages. Hence the effective current value is determined by the mobility of the ions. As the applied voltage is increased, the fraction of it involving the cathode potential drop is diminished. This stems from the fact that the field of the layer of positive charges that forms near the cathode brings about the necessary emission of electrons. Then we can say that the current in the discharge is transported by electrons having the drift velocity that corresponds to the applied potential. The potential values that mark the sharp change in slope in the voltage-current characteristics permit one to determine experimentally the cathode drop. We see from the presented results that this value is close to the corresponding calculated values.

The discussed mechanism for establishment of the cathode layer in a non-self-sustaining discharge is also confirmed by direct comparison of the calculated and experimental voltage-current characteristics (Fig. 8) of a non-self-sustaining discharge in molecular nitrogen at atmospheric pressure.[31] Here the calculated voltage-current characteristics were constructed by solving the system (26) with the boundary conditions:

$$n_e(0)\,v_e(0) = -\gamma_i n_i(0)\,v_i(0), \qquad \int_0^d E(x)\,dx = U.$$

Here d is the interelectron distance, and U is the value of the applied potential. In Fig. 8, the solid line shows the experimental voltage-current characteristic. From its slope in the high-current region, one can estimate the concentration of electrons in the gas-discharge plasma in terms of the known[60] drift velocity $v_e(E/P)$ of the electrons. This estimate gives $n_e \simeq 5 \times 10^{11}$ cm^{-3}, which agrees approximately with the rate of production of electrons by the external ionizer $S \approx 5 \times 10^{16}$ cm^{-3} sec^{-1}. The calculated voltage-current characteristic for this value of S is shown in Fig. 8 by the dotted line.

Thus the results of the studies analyzed above have shown that the size of the cathode drop amounts to a small fraction of the total potential of the gas discharge with an interelectrode spacing of the order of several centimeters when the level of the energy-input power density in the non-self-sustaining discharge is lowered (to values ~ 1 kw/cm^3 · atm).

The next part of the studies to determine the electric and gas-dynamic parameters that are required to maintain steady-state non-self-sustaining discharges in large volumes of dense gases are the studies on stability of burning of a uniform non-self-sustaining discharge.

B. Mechanisms of instabilities that develop in the non-self-sustaining gas discharge

The uniform burning of a non-self-sustaining discharge in the mixtures of molecular gases that are used in CO_2 lasers can be disrupted by an entire set of instabilities. Their development converts the discharge into an arc that is sharply nonuniform throughout the volume.

Naturally, the overheat-ionization instability that was treated in the previous section, which is due to ejection of the heated gas from the developing current filament, can also develop in a non-self-sustaining discharge.[4b,44,46,47] Here the expressions (9a) and (16) derived for the increment of this instability can also be applied to the plasma of a non-self-sustaining discharge by substituting the appropriate value of the numerical coefficient $a \equiv d\ln\sigma/d\ln N$. In a non-self-sustaining discharge this coefficient is smaller and accordingly the characteristic development time of instability is greater. This is because the ionization of the electrons in the volume arises from the external ionizer, owing to the condition (25). Hence it depends but weakly on the parameter E/N.

Yet we note, if the non-self-sustaining discharge has parameters such that the inequality (25) does not hold very strongly, that the increment of development of overheat-ionization instability will actually not differ from the value for a self-sustaining discharge. The reason for this is that the characteristic time of development of instability is not determined by the expressions (9a) and (16), but by the instant of time at which the ejection of the gas from the filament, and correspondingly, the increase in the local parameter E/N will convert the discharge at this site into a self-sustaining regime of burning. Naturally, this instant of time, which is determined by the characteristic time of variation of the ionization coefficient $\alpha(E/N)$, coincides with the time of development of instability in a self-sustaining discharge.

As we have noted, the expressions (9a) and (16) for the increment of development of overheat-ionization instability were derived under the assumption of instantaneous conversion of the Joule energy into heat. This condition is rather often not satisfied for a discharge in CO_2–N_2–He gas mixtures, owing to the slow relaxation (VT-relaxation) of the energy stored in the vibrations of the N_2 and CO_2 molecules. A treatment of the development of overheat-ionization instability of a non-self-sustaining discharge in this case that is analogous to that carried out in Chap. 2 for the self-sustaining discharge has been presented in Refs. 64 and 65. The problem was solved in these studies by linearizing the perturbations of the physical quantities with respect to the non-steady-state background of the developing discharge.

Yet we note that one can find a simple solution for a developing non-self-sustaining discharge in the treated case that demonstrates the explosive nature of the development of overheat-ionization instability, and which permits one to find its increment. Let us write down the equations that describe the development of a spatially uniform non-self-sustaining discharge by assuming that all the Joule energy is converted into the energy of vibration of molecules, which then relaxes in the time τ_{VT} into heat:

$$\rho \frac{d\varepsilon}{dt} = \sigma E^2 \tag{28}$$

$$\frac{N}{\gamma - 1} \frac{dT}{dt} = \frac{\varepsilon}{\tau_{VT}(T)}; \tag{29}$$

here ε is the specific density of vibrational energy of the molecules. We assumed in writing Eq. (28) that the characteristic time of the studied process is small in comparison with the time τ_{VT}. Therefore the fraction of the Joule energy that has been converted into heat is small in comparison with the energy stored in the vibrations of molecules.

The vibrational-translational relaxation time in the molecular mixtures that are used in CO_2 lasers is a strongly declining function of the gas temperature.[49] It can be approximated by the relationship $\tau_{VT} \sim T^{-b}$, where $b \approx 3-10$. Upon making this approximation and taking account of the fact that the conductivity of the plasma does not depend on the temperature of the gas, we can easily solve the system (28)–(29) and find the $T(t)$ relationship:

$$T(t) = T(0)\left[1 - \frac{(b-1)(\gamma-1)\sigma E^2 t^2}{2P\tau_{VT}(0)}\right]^{-1/(b-1)}. \tag{30}$$

Hence we see that the heat release in a non-self-sustaining discharge under the studied conditions is explosive in nature. In the course of a finite time of the order of:

$$\tau_T \approx \sqrt{\frac{P\tau_{VT}(0)}{b\sigma E^2}} \tag{31}$$

all the stored energy is converted from molecular vibrations into heat.[6)] Since we are dealing with discharge parameters that make the time of Joule energy release small in comparison with the time of VT-relaxation that corresponds to the initial gas temperature ($P/\sigma E^2 \ll \tau_{VT}(0)$), we see from (31) that the value of τ_t lies in the range $P/\sigma E^2 < \tau_t < \tau_{VT}(0)$. On the other hand, as we have seen above, overheat-ionization instability develops in the heat release in a non-self-sustaining discharge with a characteristic time $\tau_{ch} \lesssim P/\sigma E^2$. Hence, when a non-self-sustaining discharge has a large VT-relaxation time, one can say that overheat-ionization instability will develop with the characteristic time τ_t that is defined by Eq. (31), and which corresponds to the instant of time by which all the Joule energy has been converted into thermal energy of the molecules.

The derived expressions for the increment of development of overheat-ionization instability in the various limiting cases imply that the duration of stable burning of a non-self-sustaining discharge will increase with decreasing Joule energy-input power. Therefore a decrease in the power of the external ionizer to the least permissible level that was studied in the last section should substantially increase the duration of uniform burning. This gives the prerequisites for bringing the non-self-sustaining discharge into a steady-state burning regime.

Let us make some estimates of the characteristic time of development of overheat-ionization instability in gases at atmospheric pressure. In N_2–CO_2–He (H_2O, H_2) gas mixtures, the time for heat release is governed by the vibrational relaxation between the N_2 and CO_2 molecules and the VT-relaxation of CO_2 molecules by He (H_2O, H_2) molecules. Upon using the known data on the rates of these processes.[49,67,68] we can find that mixtures relatively strongly enriched in CO_2 and He (e.g., a mixture $N_2 : CO_2 : He = 1 : 2 : 3$) show a time for heat release of $\sim 10^{-5}$–10^{-6} sec. Yet the characteristic time of heat release is increased by factors of hundreds in mixtures that consist mainly of nitrogen alone with small (percent) additions of the other gases, owing to the extremely slow VT relaxation of molecular nitrogen.[49]

As we have seen, the characteristic time of development of overheat-ionization instability in a non-self-sustaining discharge in the case of fast VT-relaxation can be estimated by Eq. (9a) as follows: $\tau_{ch} \lesssim P/\sigma E^2$. Upon substituting the value $\sigma E^2/P \simeq 10^3$ W/atm·cm^3, we get $\tau_{ch} \lesssim 10^{-4}$ sec. Since this time is substantially longer than the characteristic energy-relaxation time in CO_2–N_2–He (H_2O, H_2) gas mixtures at atmospheric pressure, we can say that the estimate made for these mixtures of the development time of overheat-ionization instability is correct. Yet in a discharge in mixtures consisting mainly of nitrogen alone, the development time of overheat-ionization instability must be estimated by Eq. (31). For a correct estimate, we must know here the amount of impurities that lead to VT-relaxation of nitrogen molecules. For example, if a percent of added water vapor exists in nitrogen at atmospheric pressure, at room temperature we have $\tau_{VT}(0) \simeq 10^{-3}$ sec. Then the estimate of the characteristic time of development of overheat-ionization instability according to Eq. (31) with $\sigma E^2/P = 10^3$ W/cm^3 · atm gives $\tau_t < 3 \times 10^{-4}$ sec.

Goluber *et al.*[69] discuss the mechanism of ionization instability of non-self-sustaining discharges caused by population of metastable electronic levels of the nitrogen molecules in the discharge process, by the increasing role of stepwise ionization, and as a consequence, by conversion of the discharge into a self-sustaining burning regime. In the non-self-sustaining discharges that are used for exciting CO_2 lasers, a metastable

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[6)] Interestingly, volume heat generation can occur explosively also in steady-state gas discharges when cooling of the gas and relaxation of the vibrationally excited molecules occur at the walls of the gas-discharge tube under the initial conditions, owing to volume VT-relaxation that increases with time.[66]

electronic level ($A^3\Sigma_u^+$) of the N_2 molecules can become populated. Its energy is about 6 eV. This level becomes populated both by direct electron impact and by cascade transitions of electrons from higher-lying electronic levels of the N_2 molecules.[70] On the other hand, the $A^3\Sigma_u^+$ electronic state of the N_2 molecules has a long spontaneous lifetime (~1 sec),[71] and it is relatively easily quenched in collisions with molecules of N_2, CO_2, He, H_2O, and H_2,[72-75] which are usually present in the gas mixtures. For example, estimates show that quenching of the metastable states of N_2 ($A^3\Sigma_u^+$) in intermolecular collisions does not occur in a non-self-sustaining discharge in molecular nitrogen at atmospheric pressure if the duration of the discharge is $\lesssim 3\times10^{-4}$ sec.

In order to describe the fundamental features of ionization instability caused by stepwise ionization of the molecules existing in the discharge, we shall briefly study a possible conversion of a non-self-sustaining discharge into a self-sustaining burning regime.[69] We shall assume that this conversion is determined by the growing role in the discharge process of stepwise ionization of molecules excited to a metastable state. We shall write the equations of balance of the electrons and metastable nitrogen molecules in a developing uniform discharge, while assuming that detachment processes are not important in the electron balance (one can reduce the allowance for detachment processes to renormalizing the electron-ion recombination coefficient[76]):

$$\frac{dn_e}{dt} = S + k_{12}n_eN^* - \beta n_e^2 - \gamma_n n_e, \tag{32}$$

$$\frac{dN^*}{dt} = \xi S + k_{01}n_eN - k_{12}n_eN^* - \frac{N^*}{\tau}; \tag{33}$$

Here S is the rate of production of electrons per unit gas volume by the external ionizer; ξS is the rate of production of excited molecules N^* by the external ionizer (the probable value of ξ is $\lesssim 1$); β and γ_n are respectively the electron-ion recombination and attachment coefficients; τ is the characteristic quenching time of the metastable N_2 molecules in intermolecular collisions; and $k_{01}(E/N)$ and $k_{12}(E/N)$ are respectively the excitation constant of metastable levels of the nitrogen molecules and the ionization constant for these levels by the electrons of the gas-discharge plasma. These constants are sharply varying functions of the ratio E/N in the region of optimal values.[57]

Let us analyze the solution of (32) and (33), while assuming that the process of establishing the electron concentration is quasi-steady-state. That is, we shall assume that the characteristic time of the discussed instability involving the population of the metastable levels substantially exceeds the time for establishing the electron concentration ($\sim 10^{-6}$–10^{-5} sec) that arises from recombination and attachment. Then we can write the $n_e(t)$ relationship from (32) in the "recombination" and "attachment" cases respectively in the following forms:

$$n_e(t) = \frac{k_{12}N^* + \sqrt{(k_{12}N^*)^2 + 4\beta S}}{2\beta}, \tag{34}$$

$$n_e(t) = \frac{S}{\gamma_n - k_{12}N^*}. \tag{35}$$

We can picture the development process in the "recombination" case as follows. At first, as long as stepwise ionization is not essential ($k_{12}n_eN^* < S$), the electron concentration is constant ($n_e = \sqrt{S/\beta}$,), while the concentration of metastable molecules is increasing. Owing to the increase in $N^*(t)$, the discharge can convert to a self-sustaining burning regime at the instant of time t_{tr}, which is approximately given by the equation:

$$k_{12}N^*(t_{tr}) = \sqrt{S\beta}. \tag{36}$$

Evidently this transformation will occur if $N^*(t)$ can increase to the value determined by Eq. (36). In turn, as we see from (33), this will happen if the following inequalities hold:

$$t_{tr} < \tau, \tag{37}$$

$$k_{12}n_eN^*(t_{tr}) < \xi S + k_{01}n_eN. \tag{38}$$

Upon taking (36) into account, we can rewrite the inequality (38) in the form

$$\sqrt{S} < \frac{k_{01}N}{\sqrt{\beta}}. \tag{39}$$

Here we get the following expression for t_n by using (33) and (36):

$$t_{tr} = \frac{\beta}{k_{01}k_{12}N}. \tag{40}$$

If we take into account the fact that the condition for the initial non-self-sustaining nature of the discharge (25) in the studied case has the form $\sqrt{S} > k_{02}N/\sqrt{\beta}$, where $k_{02} \equiv \alpha v_e/N$ is the ionization rate constant of the molecules from the ground state, while $k_{02} \ll k_{01}$, then evidently, a broad range of powers of the external ionizer and of ratios E/N exists in which the inequality (39) is satisfied in the non-self-sustaining discharge. Here it satisfied all the more in the region of minimum possible values of the power of the external ionizer that was treated in Sec. A of Chap. 3. Then we can see from Eqs. (33) and (34) that when the time t_{tr} has elapsed from the onset of the discharge, that is, when the discharge has converted into a self-sustaining regime of burning, the concentration of the excited molecules and that of the electrons will increase as follows in time:

$$N^*(t) \sim n_e(t) \sim e^{t/t_{tr}}. \tag{41}$$

Interestingly, the characteristic time of growth t_{tr} of the electron concentration as defined by Eq. (40) in the studied "recombination" case is determined solely by the ratio E/N, owing to the strong dependence of $k_{01}(E/N)$ and $k_{12}(E/N)$, and it does not depend on the power of the external ionizer.

Upon carrying out an analogous treatment in the case in which the loss of electrons in the volume is due to attachment (such a situation can happen at not very large values of the power of the external ionizer),[31] we can

derive from Eqs. (33) and (35) the following relationship for the development of the electron concentration in time:

$$n_e(t) = \frac{S\gamma_n}{\sqrt{1-(t/t'_{tr})}}, \quad (42)$$

where

$$t'_{tr} = \frac{\gamma_n^2}{2k_{01}k_{12}NS}. \quad (43)$$

Thus we see that the development of $n_e(t)$ in this case is explosive in nature, with a characteristic time determined by Eq. (43).

The nature of the studied solutions for the $n_e(t)$ relationship in the transition of a non-self-sustaining discharge into a self-sustaining regime of burning permits us to draw the following conclusions. We see from the derived expressions (40) and (43) for the time of sharp increase in the electron concentration, and correspondingly, in the discharge current density, that a small nonuniformity in the parameters of the discharge will lead to strong nonuniformity, and hence also to instability of the course of the gas discharge. This nonuniformity can arise from heat release, which leads to weak perturbations in the gas density and correspondingly to far greater perturbations in the coefficients k_{01} and k_{12}.[77] In the "attachment" case, moreover, the initial nonuniformity can be introduced by spatial nonuniformity of the power of the external ionizer.

Estimates of the characteristic time of development of the studied nonuniformity are very difficult, owing to the lack in the literature of reliable data on the variations of the coefficients $k_{01}(E/N)$ and $k_{12}(E/N)$ in different gas mixtures. We can say qualitatively that the increment of the development of ionization instability in a non-self-sustaining discharge arising from excitation of metastable levels depends exponentially strongly on the ratio E/N in the discharge. Therefore one can always choose a value of the electric field intensity in a non-self-sustaining discharge below which the increment of this instability will be smaller than the increment of development of overheat-ionization instability. In such a case, the latter will limit the time of uniform, stable burning of the discharge. The estimates given above of the characteristic quenching time of excited states into the metastable state of nitrogen molecules indicates the possible development of the studied ionization instability at the corresponding, not very small values of the ratio E/N.

We note that the development of ionization instability of non-self-sustaining discharges arising from population of metastable electronic levels of N_2 molecules in the discharge process and from the increasing role of stepwise ionization can be exacerbated by another factor. This is the growth in time of the coefficient of self-sustaining ionization caused by the gradual population of the vibrational levels of the molecules and the concomitant decline in the fraction of inelastic losses by the electrons.[50,51] Napartovich *et al.*[76] report a numerical study of ionization instability of a non-self-sustaining discharge in molecular nitrogen involving stepwise ionization and the dependence of the rate constants of the reaction in the vibrational temperature.

The mechanism of "chemical" instability has been discussed in Ref. 79a. This can develop in the plasma of the non-self-sustaining discharges used for exciting CO_2 lasers. This instability arises when the loss of electrons is caused by their attachment to CO_2 or O_2 molecules, and any sort of excited atoms or molecules arise in the plasma that can react with the negative ions so as to detach an electron from an electronegative molecule. In the studied discharges, these active molecules can be nitrogen molecules excited into long-lived vibrational and electronic states.[80] Here instability stems from the fact that the increased concentration of electrons causes an increased concentration of chemically active excited atoms or molecules. In turn, this leads to enhanced detachment, and thus to an increased electron concentration. This feedback governs the sharp growth in the electron concentration. Just as in the case of stepwise ionization, the latter can develop nonuniformly, owing to development of perturbations in the gas density caused by nonuniform heat release.[77]

Ionization instability of a non-self-sustaining discharge, which leads to domain formation, has been studied in Refs. 46, 53, 81, 82, and 86a. The reason why this instability arises is the decline in the concentration of electrons as their temperature rises in the case in which the loss of electrons is governed by dissociative attachment of electrons to CO_2 molecules. While this instability does not convert the discharge into an arc, its development disrupts the uniform excitation of the gaseous medium. This substantially restricts the possible use of a discharge having domain structure to excite CO_2 lasers. We note that spatial nonuniformity of the ionizing radiation can analogously impair the optical characteristics of gaseous media.[83,84]

We note in concluding this section that the mechanisms of the above-discussed instabilities, whose development disrupts the uniform burning of the non-self-sustaining discharge, have characteristic increments of about the same order of magnitude. Yet a more exact determination of the development time of any given ionization instability requires an exact knowledge of the kinetic coefficients as functions of the parameters of the gas discharge, the chemical composition of the gaseous medium, the gas temperature, the degree of excitation of the electronic and vibrational levels of the atoms and molecules existing in the discharge, etc. Nevertheless, the qualitative aspect of the performed studies indicates, when other conditions are equal, that the increments of development of ionization instabilities decrease with decreasing electric power density released in the non-self-sustaining discharge.

C. The stationary non-self-sustaining discharge in gases at elevated pressure

The above-discussed possibility of substantially increased time of uniform burning of a non-self-sustaining discharge by decreasing the electric power density released in the discharge has been confirmed experi-

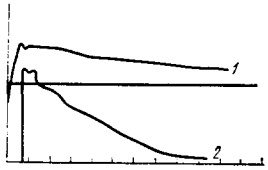

FIG. 9. Oscillograms of the current of the IFP-20000 lamp (1) and the current of the non-self-sustaining discharge (2). t = 100 μs/division; $E/P = 6.7$ V/cm·Torr.[61]

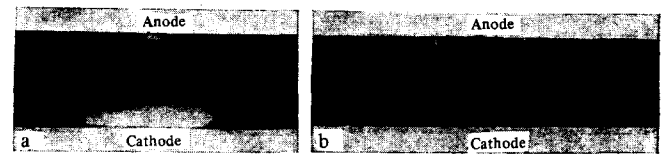

FIG. 11. Photographs of a discharge in a mixture $N_2 : CO_2 = 4$; 1 at atmospheric pressure with 0.5 sec exposure.[31] a) Stable regime; b) unstable regime.

mentally in Refs. 61, 62, 85–90. These studies used external ionization sources (ultraviolet[61,87,88]; a nuclear reactor[85]; high-energy electron beams).[62,86,89,90] They showed that a non-self-sustaining discharge can burn stably and uniformly in CO_2–N_2–He mixtures at a pressure close to atmospheric up to times of $\sim 10^{-4}$–10^{-3} sec., provided that the power density of the Joule energy input is maintained at the level: $\sigma E^2/P \sim 10^3$–10^4 W/cm^3·atm.

Figure 9 shows an oscillogram of the current in a non-self-sustaining discharge in molecular nitrogen at a pressure of $P = 500$ Torr.[61] The decline in current arises from the cessation of the discharge in the pulsed xenon lamp that served as the ultraviolet source. We see that the characteristic pulse duration of the non-self-sustaining discharge is of the order of ~0.5 μs. This range of times of stable burning of a non-self-sustaining discharge ($\sim 10^{-3}$–10^{-4} s) permits one rather simply (essentially by subsonic pumping of the gas through the working volume) to effect a gas change in the discharge chamber, and thus to remove the accumulating perturbations and to convert to a steady-state non-self-sustaining discharge at elevated gas pressures.

Steady-state burning of non-self-sustaining discharges in CO_2–N_2 gas mixtures at atmospheric pressure was first achieved in Ref. 31 in the apparatus shown schematically in Fig. 10. The authors used as the external ionizer a steady-state electron beam that was formed by an electron gun, and which entered the gas-discharge chamber through an aluminum-foil window. It had an energy of ~100 keV with a current density up to 50 μA/cm^2. The latter value is about 10^3–10^4 times smaller than the corresponding current densities of the electron beam that had been used in Refs. 3–9. Such small values of the current density of the electron beam permitted them, first, to effect steady-state transmission of the beam through the foil, and second, to maintain an energy-input power in the gas discharge at a rather low level ($\sigma E^2 \sim 10^3$ W/cm^3). They pumped the gas through the interelectrode volume at a speed of 10–100 m/sec to remove the accumulating perturbations.

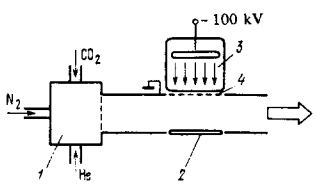

FIG. 10. Diagram of the experimental apparatus in which the steady-state non-self-sustaining discharge was studied.[31,79] 1—gas-mixing chamber; 2 and 4—cathode and anode of the discharge chamber; 3—electron gun. The arrow indicates the direction of gas flow.

Figure 7 shows the experimental voltage-current characteristics obtained in this study. The curves break off on the side of high current and voltage values in the region of unstable burning, where the discharge converts from a volume discharge into an arc discharge localized in a narrow channel.

Figure 11 shows photographs obtained in Ref. 31 of the the luminescence of a discharge in a direction transverse to the flow of the gas in a stable (a) and an unstable (b) burning regime. The glowing region near the cathode, whose thickness depends on the power input into the discharge and the velocity of gas flow, reveals the overheating of the gas in the zone near the cathode, which arises from the large electric field intensity in this region. The results of experimental study of analogous steady-state non-self-sustaining discharges in CO_2–N_2–He (H_2) gas mixtures at elevated gas pressure are given in greater detail in Ref. 79. The latter study was concerned with the relationship between the limiting energy inputs into the gas and the current density of the high-energy electrons, the rate of gas pumping, and the composition of the gas mixture. This study gave the results of studying a steady-state non-self-sustaining discharge in gas mixtures containing varying amounts of oxygen: technical-purity nitrogen—less than 0.5% oxygen; nitrogen containing 3% oxygen; and air (20% oxygen). These same mixtures were studies with additions of 7% CO_2 and 30% helium. The rate of flow of the gas in the discharge was varied from 30 to 150 m/sec. This corresponded to a variation in the time of flight of a gas molecule through the discharge gap from 1000 to 200 μsec. The gas pressure was varied from 75 to 300 Torr. Figures 12 and 13 show the results obtained in Ref. 79 from processing the experimental voltage-current characteristics, as taken at different electron-beam currents. Figure 12 shows the limiting values of the total field strength across the discharge beyond which arcs are formed in the discharge. Figure 13 shows the corresponding values of the energy input density into the discharge for a gas flow velocity of 3 m/sec and gas pressure of 150 Torr for different gas mixtures.

As we see from these diagrams, the limiting field strengths across the discharge vary for different mixtures (the limiting field substantially increases with increasing oxygen content), but they vary weakly with

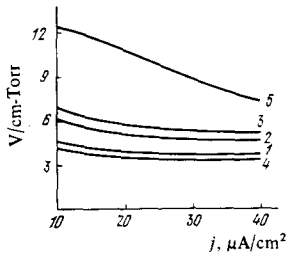

FIG. 12. Relationship of the limiting field intensity of a non-self-sustaining gas discharge to the current density of the electron beam. 1—technical-purity nitrogen; 2—nitrogen containing 3% oxygen; 3—nitrogen containing 3% oxygen and 72% CO_2; 4—$N_2 : He : O_2 = 60 : 30 : 2$; 5—air.[79]

varying current density of the electron beam. Conversely, the limiting currents, and hence also the values of the energy-input power, increase with increasing current density of the electron beam. Experimentally, the maximum energy input density at a pressure of 150 Torr, time of flight of the cold gas through the discharge chamber of 1 μsec, and maximum current density of the electron beam of 40 $\mu A/cm^2$ amounts to ~0.12 J/cm^3 for technical-purity nitrogen, ~0.1 J/cm^3 for this same nitrogen with 7% CO_2, ~0.08 J/cm^3 for nitrogen with 3% oxygen, ~0.06 J/cm^3 for nitrogen containing 3% oxygen and 8% CO_2, and ~0.04 J/cm^3 for air. Addition to the mixture of up to 30% helium does not substantially change the voltage-current characteristics of the discharge and the limiting energy inputs. Moreover, as was shown in Ref. 79, the limiting energy input per unit gas volume increases proportionally with increasing pressure. Yet the limiting energy input density falls slightly with increasing gas-flow velocity, in line with the fact that the limiting value of E/P for a stable burning regime increases to a lesser degree than the gas-flow velocity. An analogous experimental result has been obtained in Ref. 90.

The results of Refs. 31, 79, and 90 permit us to draw two fundamental and essential conclusions: 1) The experimentally realized steady-state values of the current density of the beam of ionizing electrons at which the energy-input power density in the discharge has declined to the level $\sigma E^2/P \sim 10^3$ W/cm^3·atm allow a steady-state, stable energy input into a CO_2–N_2–He gas mixture at the level of 0.3–0.5 J/cm^3·atm. 2) There is a threshold value $(E/P)_{max}$ in the studied gas mixtures above which the gas discharge transforms into an arc. As we have noted, the value of $(E/P)_{max}$ is determined mainly by the composition of the gas mixture, and it depends weakly on the intensity of the ionizing radiation and on the speed of gas flow. At ratios E/P below this threshold value, the limiting energy input into the discharge that is attained in such a case by increasing the power of the external ionizer or by reducing the speed of gas flow is approximately constant ($(\sigma E^2/P)t \simeq$ const, where t is the time of flight of a gas molecule through the excitation zone). The existence of such a threshold value of E/P has also been noted in Refs. 85, 61, and 62, which were concerned with studying pulsed quasi-steady-state non-self-sustaining discharges in gases at elevated pressures.

The observed nature of the development of instabilities, which restricts the energy input in a steady-state non-self-sustaining gas discharge, fits qualitatively into the overall pattern discussed above of the development of ionization instabilities, in which the stability of burning of the discharge at low values of the ratio E/P is primarily restricted by the development of overheat-ionization instability. When the threshold value of E/P is exceeded, the gas discharge is observed to convert sharply into a self-sustaining burning regime. The latter is caused by the perturbations in the initial ionization characteristics of the medium that accumulate along the flow of gas. However, as we have noted above, it takes a complex diagnostics of the gas-discharge plasma and exact calculation of the most important kinetic processes in order to single out definitely any given mechanism of instability.

We should note as another process that also affects the stability of uniform burning of a non-self-sustaining discharge the breakdowns and formation of localized arcs in the layer next to the cathode. When the cathode fall is a substantial fraction of the total voltage applied to the gas-discharge gap, these can lead to breakdown of the entire gas discharge, and thus they can restrict the limiting energy inputs into the steady-state non-self-sustaining discharge to a lower level than that indicated above.[79,89] The authors of Ref. 79 were able substantially to suppress the instabilities in the region next to the cathode in a non-self-sustaining gas discharge by overheating, with consequent ejection of the gas from the zone next to the cathode, or by mixing pure helium into this zone. This effect is explained by the increased rate of multiplication of electrons in the cathode layer, and thereby by the decreased cathode drop that is needed to maintain the proper rate of emission of electrons from the cathode layer. Moreover, the decline in the gas density in the layer next to the cathode or the replacement of the gas mixture in this layer by helium alone substantially decreases the normal current density at the cathode, and thus it leads to a possible transition from a subnormal to a normal burning regime at the cathode. As we know,[60] the latter substantially stabilizes the uniform covering of the cathode with the flowing gas-discharge current. In this regard we note Refs. 91 and 92, in which a rarefaction of the gas in the region next to the cathode of self-sustaining discharges also stabilized the uniform burning of the discharge.

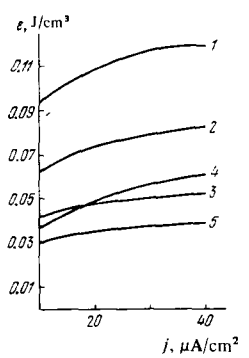

FIG. 13. Relationship of the limiting energy input density in a non-self-sustaining gas discharge to the current density of the electron beam in different gas mixtures.[79] (The numbering of the curves follows Fig. Fig. 12.)

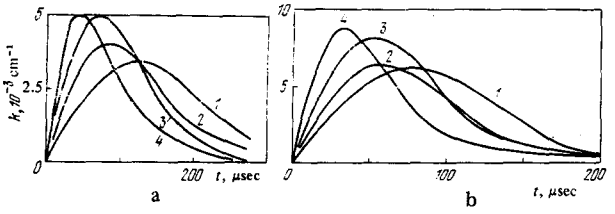

FIG. 14. Calculated time course of the amplification coefficient: a) at a pumping power of 1 kW/cm^3, composition of the N_2–CO_2–He mixtures: 1–85 : 5 : 10; 2—8 : 1 : 1 : 3—6 : 1 : 3; 4—5 : 2 : 3; b) at a pumping power of 2 kW/cm^3, composition of the N_2–CO_2–He mixtures: 1—65 : 5 : 30; 2—8 : 1 : 1; 3—6 : 1 : 3; 4—5 : 2 : 3.[86]

Thus the above-discussed results of the studies have proved that one can design in practice systems in which a steady-state non-self-sustaining discharge is maintained in gases at elevated pressures. As we have noted, it has proved possible to convert from a pulsed to a steady-state burning regime of a non-self-sustaining discharge upon considerably reducing the power density developed in the discharge. Naturally, such a power reduction leads to a corresponding decrease in the rate of excitation of CO_2 molecules to the upper laser level.

Hence the next step in the studies needed for design of CO_2 lasers having an elevated pressure of the working medium was to study the optical properties of dense gases when excited by a steady-state non-self-sustaining discharge.

4. OPTICAL CHARACTERISTICS OF GASEOUS MEDIA BASED ON CO_2 WHEN EXCITED BY A STEADY-STATE NON-SELF-SUSTAINING DISCHARGE

The kinetic processes that govern the inverted population of the CO_2 molecules in gaseous media when excited by a glow discharge have been treated in detail in many studies (see, e.g., the reviews).[49,67,93] They can be summarized as follows.

The source for creating the inverted population of CO_2 molecules in CO_2 lasers is the electrons of the gas-discharge plasma, whose energy almost completely goes into exciting the upper laser level of the CO_2 molecules. Yet the relaxation of the upper laser level of the CO_2 molecules is caused by intermolecular collisions whose probability strongly increases with increasing gas temperature. The population of the lower laser level of the CO_2 molecules arising from fast processes of vibrational-translational relaxation in intermolecular collisions is close to the equilibrium value corresponding to the gas temperature. Therefore it is precisely the overheating of the gas that restricts the limiting energy input into the medium to the values at which both the concentration of CO_2 molecules existing in the lower laser state and the rate of relaxation of the upper laser level become large enough to abolish the inverted population of CO_2 molecules.

Since the rate of excitation of CO_2 molecules to the upper laser level in a gaseous medium being excited by a steady-state non-self-sustaining discharge is substantially smaller than the corresponding values that are characteristic of pulsed discharges, the limiting gas temperature should be reduced in the case of steady-state excitation. Correspondingly the limiting energy input density into the gas at which the population of CO_2 molecules is inverted should also be reduced. Simple estimates of the relationship of the duration of laser action to the electric energy input power, and correspondingly, an estimate of the limiting energy input into the gas are highly difficult, owing to the strong temperature-dependence of the rates of relaxation of the laser levels of CO_2 molecules.[49] Therefore we shall give the results of a numerical calculation of the time course of the amplification coefficient in CO_2–N_2–He at atmospheric pressure obtained in Ref. 86 with account taken of the increase in the gas temperature during the pulse. From this we shall obtain the theoretical characteristics that govern the limiting energy input into the medium at the values of the Joule energy-input power that characterize steady-state non-self-sustaining discharges in dense gases.

Figure 14 shows the results of calculating the time-dependence of the amplification coefficient in CO_2–N_2–He gas mixtures differing in partial composition at atmospheric pressure for electric-energy-input powers in the gas discharge of 1 kW/cm^3 and 2 kW/cm^3. We see from these diagrams that the duration of existence of the inverted population of CO_2 molecules amounts to ~200–300 μsec, so that the studied level of pumping powers permits one to get an energy input of ~0.3–0.4 J/cm^3. As was noted in the last section, about the same level of energy inputs is restricted also by the development of ionization instabilities in the plasma of steady-state non-self-sustaining discharges.

With further reduction of the energy-input power into the gas discharge, the inverted population of CO_2 molecules disappears at a lower overheating of the gas, and hence at lower energy inputs than those that one can attain in a stable burning regime of a non-self-sustaining discharge. The tendency to such a decline is quite visible from comparing the results shown in Fig. 14 for different values of the pumping power. Hence we can say that the burning regime studied in Refs. 31, 79, and 90 of steady-state non-self-sustaining discharges is optimal for exciting continuous-wave CO_2 lasers operating at elevated gas pressures.

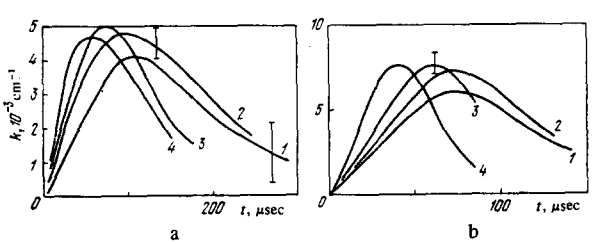

FIG. 15. Time course of the amplification coefficient: a) at a pumping power of 1 kW/cm^3, composition of the N_2–CO_2–He mixtures: 1—8 : 1 : 1; 2—6 : 1 : 3; 3—4 : 1 : 5; 4—5 : 2 : 3; b) at a pumping power of 2 kW/cm^3, composition of the N_2–CO_2–He gas mixtures: 1—8 : 1 : 1; 2—6 : 1 : 3; 3—4 : 1 : 5; 4—5 : 2 : 3.[86]

Steady state laser action of CO_2 lasers (or quasi-steady-state at an analogous level of pumping power) excited by a non-self-sustaining discharge in gases at elevated pressures has been obtained experimentally in Refs. 79b, 85–89, and 94–96. In Ref. 86, the experimental variation of the time course of the amplification coefficient was obtained in gas media differing in partial composition at atmospheric pressure when excited by a quasi-steady-state non-self-sustaining discharge. A beam of high-energy electrons was used as the external ionizer with a current density of ~100 $\mu A/cm^2$, which is characteristic for creating conditions for conversion to a steady-state regime of burning of the discharge.[31] Figure 15 gives the experimental results at an electric pumping power of 1 and 2 kW/cm^3, and they agree well with the corresponding calculated values (see Fig. 14). It has been noted in Refs. 95 and 96 that the use of non-self-sustaining discharges in the studied regimes of exciting CO_2 lasers permits replacing the helium, which is usually present in substantial amount in the working gas composition, by a small amount (~1–2%) of hydrogen. This replacement has no substantial effect on the electric-discharge characteristics nor on the optical characteristics.

5. CONCLUSION

To summarize the discussion given above, we can say that the recent results on studying non-self-sustaining discharges in molecular gases have permitted attaining a steady-state burning regime of glow discharges at elevated gas pressures (of the order of a hundred Torr and higher). These discharges are maintained over a broad range of electric-field intensities and gas pressures. Consequently they are highly promising in the applications where people usually use steady-state low-pressure gas discharges. One of the applications that we have discussed in detail in this review is the use of steady-state non-self-sustaining discharges for designing continuous wave CO_2 lasers that operate at elevated gas pressures. Naturally, also other gas lasers (e.g., CO lasers) can be excited by the steady-state non-self-sustaining discharges discussed in this review. A natural application of such discharges is also to use them as a steady-state plasma-chemical reactor.

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