High-pressure volume discharges in gas lasers

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Investigations of pulsed volume discharges at pressures $p \sim 1$ atm in nitrogen, CO_2:N_2:He mixtures, and mixtures of noble gases and halogens are reviewed from the point of view of their use in lasers. The review examines self-sustaining discharges in which the initiating electrons are produced by illuminating the gap with an auxiliary spark, with an x-ray source, etc., and non-self-sustaining discharges with external gas ionization by fast electrons. Data on the contraction of volume discharges are presented. A special section is devoted to high-pressure discharges in CO_2 lasers.

CONTENTS

1. INTRODUCTION

In the first gas lasers the active media were excited with the use of a steady-state glow discharge, which, as is well known, burns stably at low gas pressures (no more than a few tens torr).[1] Interest in active media based on a glow discharge increased markedly after the invention in 1964 of the cw CO_2 laser.[2] Because the lasing occurs on the low-lying vibrational-rotational transitions of the electronic ground state of the CO_2 molecule, and because of the efficient mechanisms for pumping the upper level and emptying the lower level, the CO_2 laser has a high efficiency. One of the most important problems is how to increase the average power and the pulse power of these lasers. It has been possible to do this by increasing the gas pressure or by using a pulsed discharge. However, when the pressure is increased, the glow discharge becomes an arc discharge.[1] While a pulsed power source has permitted a certain pressure increase, in order to attain a radiated energy of 5 J at 200 kW peak power very high voltage pulses (10^6 V) were required.[3]

After the appearance of Ref. 3, it became quite clear that fundamentally new concepts for pumping the active media were needed. The first lasers, including the CO_2 laser, used the so-called longitudinal glow discharge, in which the current flowed along the axis of the resonator. An important step in the development of the technology of gas lasers was the creation of a transverse excitation system, in which the electric field and the discharge current are transverse to the axis of the resonator.[4] In pulsed transverse excitation of an N_2 laser[4] emission was obtained at a peak power of 200 kW at a wavelength 3371 Å, with a nitrogen pressure of 20 torr. The pulsed source and the transverse excitation system immediately led the investigators to see the necessity of increasing the peak power of the CO_2 laser and the radiated energy by increasing the gas pressure.

An important event in the solution of this problem was the advent of open volume discharges at high pressure. Here, high pressure means a pressure that is considerably above that of a glow discharge and approaching and exceeding atmospheric pressure. Some investigators mistakenly believe that the idea of a high-pressure volume discharge grew out of work on gas lasers. As we shall show in this review, the ideal of self-sustaining and non-self-sustaining high-pressure volume discharges, and the experimental investigations of them appeared in the physics of gas discharges prior to the time that they were used in gas lasers.

The fundamental idea that made it possible to realize a self-sustaining high-pressure volume discharge was the idea of multielectron initiation, which was described and developed first in 1966.[5] The idea is this: before applying the voltage pulse across the gas discharge gap, a uniform initial concentration of initiating electrons is produced in the gap, this initial concentration being necessary in order to obtain a large discharge current as a result of the simultaneous development of a large number of electron avalanches. This idea is used in all gas lasers that operate with a self-sustaining high-

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pressure discharge, and these have been called TEA lasers. In the first of these[6] the multielectron initiation was accomplished by means of ultraviolet illumination from an auxiliary discharge, the current of which was confined by an insulating barrier. All subsequent TEA lasers operating at or above atmospheric pressure differ from that of Ref. 6 only in the systems used for initiating the electrons and for storing up the energy.[7] In the self-sustaining volume discharges the electric field turns out to greater than necessary for optimal pumping of the vibrational levels of the N_2 and CO_2 molecules. This has been pointed out in Ref. 8, where a beam of protons was injected into a tube containing a glow discharge, and because of the increase in conductivity, the burning voltage was decreased and the emission power output was increased. A non-self-sustaining discharge utilizing ionization of the gas by an electron beam was suggested and implemented in Ref. 9, and the first report on its use in CO_2 lasers for the generation of radiation, appeared in Ref. 10, and for amplification in Ref. 11. Prior to the work of Refs. 10 and 11, it was proposed[12] that for pulsed CO_2 lasers operating at low fields the free electrons be produced by adding cesium vapor to the gas and ionizing it with ultraviolet light.

System for pumping based on high-pressure volume discharges, first used for CO_2 lasers, were later also used for other gas lasers. There have been a very large number of papers published on this subject. For instance, in a review[7] published in 1974 about 700 references were cited, and laser operation at more than 500 wavelengths was reported. It is natural that in the more recent years, after the appearance of work on excimer lasers,[13,14] chemical lasers,[15] pulsed-periodic CO_2 lasers,[16] CO lasers,[17] etc., the number of publications has increased many times. It is understandable that in this review it is not possible to give a complete account of these investigations. Therefore, we shall discuss mainly the physics of high-pressure volume discharges, and only partially CO_2 lasers pumped by these discharges.

2. SELF-SUSTAINING VOLUME DISCHARGES

2.1 The role of multielectron initiation in a volume discharge

At gas pressures p of the order of atmospheric and above and at electric fields greater than the breakdown field, a streamer discharge develops.[18] For this discharge to develop, only a single initiating electron is sufficient, and it leads to the formation of an electron avalanche with a critical number N_{cr} of charge carriers. Thereafter a streamer and a spark channel are formed. The formative time of the breakdown is given by the relation

$$t_F = (\alpha v)^{-1} \ln N_{cr}, \tag{1}$$

where α is the impact ionization coefficient and v is the electron drift velocity.

The streamer breakdown mechanism has been demonstrated experimentally by applying to a gas-filled gap an overvoltage Δ of some tens of percent.[18] In Ref. 19 t_F in air was measured for $\Delta \gtrsim 100\%$. For initiation of the first electrons the gap was illuminated from an auxiliary spark. The experimental time t_F and that calculated from formula (1) were in agreement, and so it was concluded that the streamer breakdown mechanism was operative. This point of view has been generally held to be correct.[18]

However, it has been shown[5] that the interpretation of the results of Ref. 19 contained a contradiction which compelled a reexamination of the conclusion concerning the breakdown mechanism. In Ref. 19 it was assumed that the observed discharge is initiated by single electrons. However, from an analysis of the investigation reported in Ref. 19, it followed that the experiment agreed with the streamer theory when the number N_0 of initial electrons is $N_0 \approx 10^4$. This contradiction made it possible[5] to give a different physical interpretation of the experiments reported in Ref. 19. It was shown that the stage of rapid current growth is not due to the transition from a streamer to a channel, but due to the current of all the electron avalanches created by the initial electrons. From this it followed at once that the discharge current is not accompanied by the formation of a channel, but can have a volume character. On the basis of this principle, equations were obtained[5] for calculating the electric field $E(t)$ and the current density $j(t)$ of the discharge. It was assumed that a voltage pulse U_0 is applied to a gap of width d from a generator along electrical lines of characteristic impedence z:

$$e n_0 v \exp \int_0^t \alpha v \, dt = j(t), \tag{2}$$

$$j(t) = [E_0 - E(t)] \frac{1}{zs}, \tag{3}$$

where s is the area of the electrodes, n_0 is the concentration of initiating electrons in the gap, and E_0 is the initial electric field. From Eqs. (2) and (3) we have

$$\frac{dE}{dt} = -E\left(1 - \frac{E}{E_0}\right) \alpha v. \tag{4}$$

Since α/p and v are functions of the ratio E/p, it is possible from Eqs. (2)–(4) to calculate $j(t)$ and $E(t)$.

To check the conclusions of the proposed theory, experiments were carried out on the breakdown of air by pulses having a leading edge width of $3 \cdot 10^{-10}$ sec.[20,21] Preliminary gas ionization was achieved with the radiation from an auxiliary spark. The number of initiating electrons was adjusted to lie in the range 1 to 10^5. A typical oscilloscope trace of the voltage across the gap for $N_0 \sim 10^4$ (Fig. 1) has three characteristic sections: the formative time t_F, a section where the voltage falls off sharply (the switching time t_s), and a section of slow falloff, t_b, which corresponds to the volume burning phase of the discharge. The experiments showed

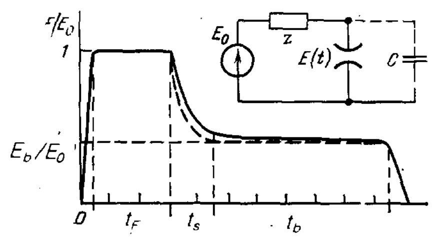

FIG. 1. Equivalent circuit and typical time dependence of field in the gap.[21] Time scale, 2 nsec, $p = 1$ atm nitrogen, $d = 0.44$ cm, $E_0 = 68.5$ kV/cm, dashed line, calculation from formulas (2)–(4).

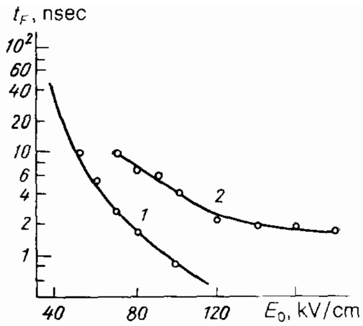

FIG. 2. Breakdown formative time as a function of electric field in air. Curve 1: results of Ref. 19; points on curve 1: results from Refs. 5 and 20 for multielectron initiation. Curve 2: same for single electron initiation.

that the time t_b depends on N_0. For $N_0 \approx 10^4$, t_b coincides with that measured in Ref. 19, while in the case of a single initiating electron the experimental time is significantly greater (Fig. 2).[5,20] From (2) and (3) it follows that

$$t_F = (\alpha v)^{-1} \ln \frac{i_F}{e n_0 v}, \tag{5}$$

where $j_b = i_b/s$ is the current density at which t_b is measured. Formally, (1) and (5) coincide for all practical purposes since the argument of the logarithm has little effect on the result. This was also the reason for the erroneous conclusion[19] that the streamer discharge mechanism was the correct one. The time t_s has an important significance in the application of the volume discharge in high-current nanosecond switches.[21,22] It can be estimated from the ratio of E_0 to the maximum slope $(dE/dt)_m$, which can be found from (4):

$$t_s \approx \frac{E_0}{(\mathrm{d}E/\mathrm{d}t)_\mathrm{m}}. \tag{6}$$

The time t_s obtained from (6) agrees with experiment.[21]

The transition of the discharge to a quasi-steady-state form (the time t_b in Fig. 1) is due to a reduction in the coefficient of impact ionization and in the electron drift velocity v as a result of a decrease in the electric field. As a result, the variation of $E(t)$ is slight and we can assume that a certain discharge field E_b is characteristic of this phase.[5,21] The role of the stage t_b in laser pumping is examined in the next paragraph of this section.

It has been confirmed by the interrupted discharge method and by electron-optical photography[23] that in nitrogen, air, CO_2, and other gases at atmospheric pressure, a multielectron-initiated discharge has a volume character (Fig. 3a). In the case of single-electron initiation one or several narrow channels are seen in the gap (Fig. 3b).[23]

Even before the first TEA lasers were developed, the self-sustaining volume discharges was used to produce nanosecond switches.[9] In this effort the effect of the inductance of the spark channel on the steepness of the current increase was successfully eliminated, and pulses with a rise time down to 10^{-10} sec were obtained. The maximum switched current was 10^5 A for an electrode area of 150 cm² and a discharge volume greater than 100 cm³. For the preliminary ionization a discharge along the surface of a ceramic with a high ε was used. This was the first instance of the use of an insulating cathode with a gliding discharge for excitation of a volume discharge in a gap.[9] Subsequently this sytem became widely used also in lasers.

2.2 Self-sustaining volume discharges in gas lasers

Thus, the general principle of igniting a pulsed volume discharge is the formation of the necessary concentration n_0 of electrons in the gap and their subsequent multiplication by ionization. Beginning in 1969 a large number of systems were devised for the excitation of CO_2 lasers where the initiating electrons were produced by illumination of the gap from auxiliary discharges of various types: a discharge with confinement of the current by an insulating barrier,[6,24] a corona discharge,[25–27] a spark,[28] and others.[7,16] An efficient source of gas photoionization was the "plasma cathode".[29,30] In Ref. 31 a plan was suggested for igniting a volume discharge with two plasma electrodes. To increase n_0 the use of an easily ionized admixture such as alkali metal vapors was suggested.[12] For this purpose organic compounds have been added to the laser mixture CO_2:N_2:He.[32] In excimer lasers the required level of n_0 is sometimes produced by ionization with x rays.[33,34] This method is a promising one for discharges in gaps where the spacing is some tens of centimeters.[35] In the work of Ref. 36, the ionization was maintained by the steady radiation of radioisotopes.

Producing the necessary concentration n_0 of the initiating electrons is one of the basic prerequisites for the formation of a volume discharge. The first estimate of n_0 was made on the basis of the condition that the increase of the current to its maximum should occur in a time that is no greater than the time of development of the avalanches to the critical size[9]:

$$n_0 \geqslant \frac{j}{e N_{cr} v}. \tag{7}$$

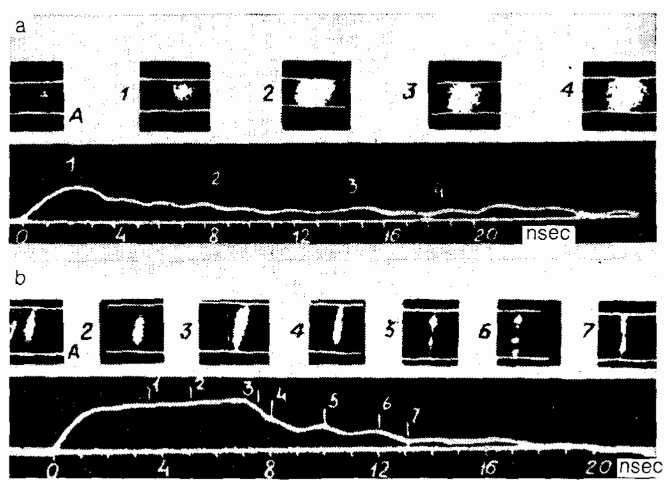

FIG. 3. Results of electron-optical photography of discharge in air for multielectron (a) and single electron (b) initiation.[23] Exposure time, 4 nsec, $p = 1$ atm. a) $d = 0.6$ cm, $E_0 = 100$ kV/cm. b) $d = 0.8$ cm, $E_0 = 73$ kV/cm.

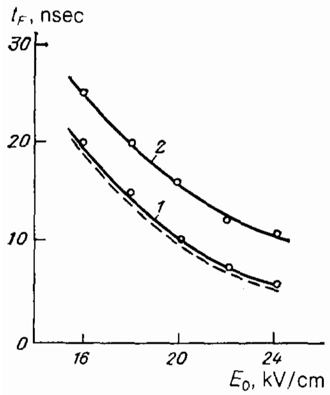

FIG. 4. Voltage dependence of breakdown formative time in argon for volume (1) and surface (2) discharge initiation.[43] Dashed line is calculation from (5).

A number of investigators[37–40] have estimated n_0 on the basis of the condition of spatial overlap of the avalanches that are generated from the initiating electrons. Overlap occurs when, after a characteristic time $t = 1/\alpha v$, the size of an avalanche is at least $n_0^{-1/3}$. Depending on the way one estimates the radius r of an avalanche, one can arrive at some differing criteria. If r is determined from the electron diffusion process as $r = (6Dt)^{1/2}$, then we obtain[37]

$$n_0 \geqslant \left(\frac{3\alpha e E_0}{32 U_T}\right)^{3/2}, \tag{8}$$

where E is the diffusion constant and u_T is the average electron energy in the avalanche. The estimate of n_0 by formula (8) or other similar formulas lead to the value $n_0 \approx 10^6$–10^8 cm^{-3}.

Experimental measurements of the average concentration of initiating electrons for specific laser designs have been made by many investigators.[16,30,37,41,42] When easily ionized organic compounds are added to the molecular gases, n_0 can reach 10^{11} cm^{-3},[16,41] and when these compounds are absent the concentration decreases by several orders of magnitude. Measurements[42] have shown that up to 80% of the total number of initiating electrons are formed as a result of the photoelectric effect at the cathode. An account of the influence of the photoelectric effect and the photoionization of the gas in the formation of the volume discharge is given in Ref. 43. Two modes of producing the initiating electrons were employed: at the cathode only, or at the cathode and in the volume of the gas simultaneously. Figure 4 shows the breakdown formative times for these two cases, and Fig. 5 shows photographs of the light emission from the discharge at various instants of time. In the case of surface initiation the density of secondary electrons is insufficient for the mutual overlapping of the secondary avalanches, and the discharge develops in the gap in the form of a large number of filamentary channels (frame 2). In the case of volume initiation a uniform discharge develops (frame 4). A calculation of t_F from formula (5) for $n_0 = 2 \cdot 10^4$ cm^{-3} (Fig. 4) agrees well with the experimental value, and this allows us to estimate the density of initiating electrons that is sufficient for the ignition of a volume discharge.

The uniformity of the discharge is improved as the rise time of the pulse is decreased.[39,44,45] If a pulse with a long rise time is applied to a gap having a concentration n_0 of initiating electrons, then at first when the voltage is small, the electrons drift towards the anode and do not participate in the ionization multiplication.[39] The region near the cathode becomes depleted of electrons and in this region conditions are created that are favorable for the development of streamer channels or a discharge structure that is similar to that shown in Fig. 5, frame 2. When the interelectrode spacings are small, $d = 1$ cm, then it is no longer possible to obtain a volume discharge even when the pulse rise time is 10^{-7} sec.[45] For gaps tens of centimeters wide this effect is less important. For example, volume discharges have been obtained[35] in mixtures of CO_2:N_2:He with gas ionization by x rays, a gap width $d = 20$ cm, and a pulse rise time 10^{-6} sec. At a discharge time of 1.5 μsec incomplete spark channels are observed near the cathode. These can be eliminated by irradiating the gap during the entire rise time of the voltage pulse.

An important characteristic of the discharge is the discharge burning voltage or the ratio of the field strength to the pressure E_b/p. It has been established experimentally for a discharge in nitrogen and in mixtures of CO_2:N_2:He that in the stage of volume burning the ratio E_b/p is maintained close to the breakdown ratio[23,46,47] and that typical values of the specific energy put into the gas before channel formation are 0.1–0.3 J/cm^3-atm,[7,16,37] and accordingly the time of the stable burning decreases with increasing j. The ratio E_b/p depends weakly on the discharge current density[48,49] and it decreases with increasing pressure.[48,50]

We shall determine the ratio E_b/p on the basis of a model of avalanche multiplication, assuming that the burning voltage corresponds to some small value of the slope dE/dt.[21] Then, it can be seen from (4) that E_b/p is independent of the characteristic impedance, depends weakly on the initial field E_0, and decreases negligibly with increasing pressure. This model is in good agreement with experiment for volume discharge times $t_b \leqslant 10^{-7}$ sec and electron concentrations $n \lesssim 10^{14}$ cm^{-3}.[51] As $E(t)$ decreases with time and $n(t)$ increases, it is necessary to take recombination processes into account. Moreover, the increase in n leads to an

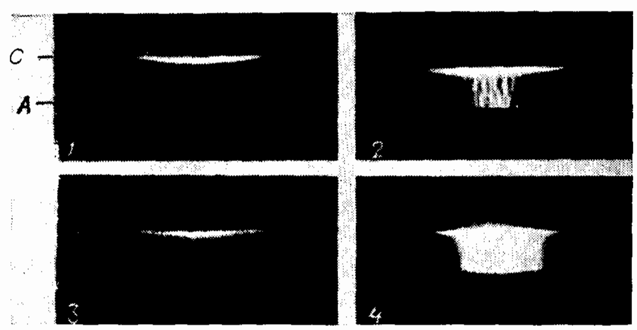

FIG. 5. Photographs of light emission in the gap during the formative stage t_F in argon for the case of electron initiation at the cathode surface (frames 1 and 2), and at the surface and in the volume (frames 3 and 4).[43] $U_0 = 18$ kV, $p = 1$ atm, $d = 1$ cm. Discharge burning time: 1) 12.5 nsec, 2) 16.5 nsec; 3) 10 nsec; 4) 12.5 nsec.

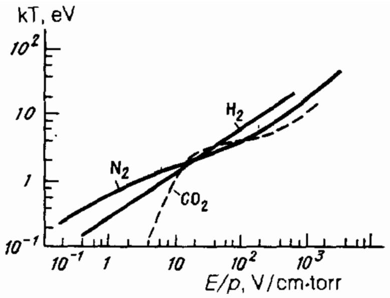

FIG. 6. Measured average electron energy in avalanches for various gases as a function of E/p.[57]

increase in the concentration of excited particles and the rate of charged particle production increases as a result of stepwise ionization.[48,49] Calculations taking into account these factors for various electrical circuits have been carried out in e.g., Refs. 46 and 52. The relative decrease in E_b/p for mixtures of CO_2:N_2:He when these factors are taken into account, in comparison to calculations carried out with the model of Ref. 5, amounts to 20–25%. However, in a discharge in helium, argon, or helium-argon mixtures, with small admixtures of halogen-containing compounds, i.e., in the case of the active media of excimer lasers, stepwise ionization processes play a governing role.[53–56] Here the burning voltage in the volume stage is several times smaller than that determined taking into account only the coefficient α.[53]

From the discussion presented above it is clear that in the breakdown of a gap to which an overvoltage is applied, E/p varies over a wide range during the formation and burning of a volume discharge. The electron energy in the plasma also varies correspondingly (Fig. 6).[57] This brings up the possibility of using a volume discharge of pumping lasers of various types. Most of the energy is deposited into the discharge during the time t_b. As a rule, it is just during this phase that the pumping of CO_2 and excimer lasers occurs. Another situation characterizes the excitation of lasers that are based on self-limited electronic-vibrational transitions of molecules (e.g., N_2 or H_2).[58] In these cases the optimal excitation occurs at high electron energy and short burning times which are limited by the lifetime of the upper level. The pumping of such systems proceeds usually under conditions of large E/p, i.e., during the portion t_s of the curve, where the voltage falls off sharply, and at the leading edge of the discharge current rise.

The development of the technology of lasers with a high-pressure self-sustaining discharge has made it possible to solve the problem of igniting these discharges in volumes of hundreds of liters, with gap widths tens of centimeters, and to bring the energy of CO_2 lasers up to the kilojoule level. Discharges with $d = 20$–30 cm have been obtained with the use of ultraviolet preionization,[27,28,59] in mixtures with organic compounds added, [60]and with the initiating electrons produced by low-energy x rays.[35] In the work reported in Ref. 61, an auxiliary generator of short pulses was used to form the discharge and then the energy from the main accumulator was fed in over a period of 2–3 μsec.

The use of self-sustaining volume discharges in lasers has certain limitations. For example, it is difficult to obtain active media at above atmospheric pressure. The burning time usually does not exceed 10^{-6} sec, and this is a result of contraction. In CO_2 and CO lasers E_b/p is greater than necessary for optimal pumping.[8] The way out of this situation was found after the discovery of the non-self-sustaining volume discharge with ionization of the gas with a beam of fast electrons.

3. NON-SELF-SUSTAINING VOLUME DISCHARGES

This discharge was produced for the first time in the work reported in Ref. 9. At a nitrogen pressure up to 15 atmospheres, a voltage considerably less than the breakdown voltage was applied to the interelectrode gap, and into this gap, through the cathode made of aluminum foil, an electron beam was injected at a current density 4 A/cm², a pulse width 20 nsec, and 400 keV energy. Under these conditions the current in the discharge reached 12 kA. These experiments were the logical extension of the work on multielectron initiation, and they were carried out for the purpose of obtain stable gap ignition and complete current during switching. A comprehensive study of non-self-sustaining discharges as applied to this problem was carried out in Ref. 62. In particular, firing of a switch was achieved at voltages up to 1 MV and currents to 40 kA.

The first application of a non-self-sustaining discharge with electron-beam gas ionization to CO_2 lasers was reported in Refs. 10, 11, and 63. In these lasers the discharge voltage was reduced and the efficiency of pumping the vibrational levels of the N_2 and CO_2 molecules was increased. This situation is illustrated by calculations, the results of which are shown in Fig. 7.[64,65] The physics and technology of CO_2 lasers immediately underwent a vigorous development. Within a few years laser energy and the volume of the active medium went from a fraction of a Joule and a few cubic centimeters to 5 kJ and hundreds of liters.[66] In the first lasers, discharges were used with current duration 10^{-7}–10^{-5} sec, and then continuous operation was achieved in a gas flow at atmospheric pressure.[68] It has been proposed that lasers based on pumping with a non-self-sustaining discharge with gas ionization by an electron beam be called

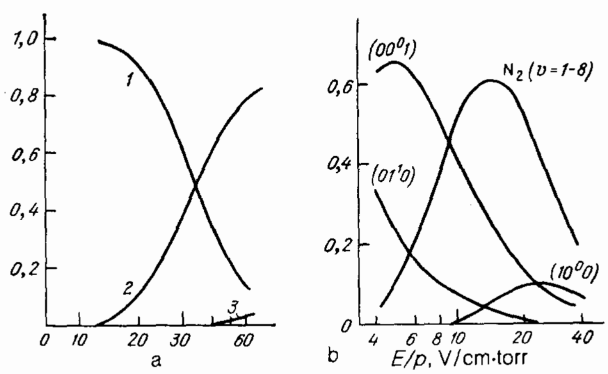

FIG. 7. Fraction of discharge energy transferred into excitation and ionization of the various levels in nitrogen (a, Ref. 64), and a mixture CO_2:N_2 = 1:1 (b, Ref. 65), as a function of E/p. 1) vibrational levels, 2) electronic levels, 3) ionization.

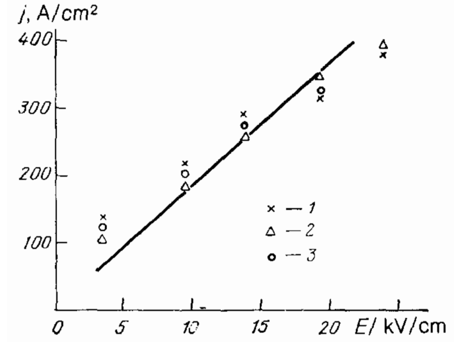

FIG. 8. Current density as a function of electric field for a non-self-sustaining discharge in nitrogen with initiating electrons from a 20 nsec beam of electrons.[62] $j_p = 10$ A/cm^2, 1) $p = 4$ atm; 2) $p = 7$ atm, (3) $p = 10$ atm.

electroionization lasers.[69] Investigations to volume discharges and their use in molecular lasers have been discussed in various reviews.[7,69–73] Special reviews have been devoted to excimer lasers.[19,55,74,75]

The main properties of current condition in discharges with gas ionization by an electron beam can be analyzed qualitatively in the following way. Let us assume that within an interelectrode gap to which a voltage U_0 is applied the volume rate of ionization is ψ and that charged particles are lost via recombination. Then the time dependence of the discharge current is found from the solution of the equations

$$\frac{\partial n}{\partial t} = \alpha v n + \psi - \beta n^2, \tag{9}$$

$$j = en\mu E, \tag{10}$$

where β is the recombination coefficient, n is the electron concentration, and μ is the mobility. Equations (9) and (10) are valid when the concentration of electrons in the discharge column is sufficiently high, $n \gtrsim 10^{11}$ cm^{-3}. Here the current in the cathode layer is maintained by impact ionization and secondary processes at the cathode. The cathode drop U_c under typical conditions of laser pumping at high pressures is much smaller than U_0, $U_c \ll U_0$, and the width of the layer $l_c \ll d$.[63,69]

When $\alpha v \ll \beta n$, i.e., in the non-self-sustaining discharge mode, we obtain from (9) the electron concentration in the discharge column as a function of time after turning on the external ionizer (the electron beam):

$$n(t) = \left(\frac{\psi}{\beta}\right)^{1/2} \mathrm{th}\,[(\psi\beta)^{1/2} t]. \tag{11}$$

The concentration $n(t)$ approaches a steady state value $n_c = (\psi/\beta)^{1/2}$ with a characteristic time $t_c = (\psi\beta)^{-1/2}$. When the beam current persists over a long period of time $t_p \gg t_c$ we have a discharge that is maintained by the beam. The specific energy of the discharge is given by the relation

$$w = e\mu \left(\frac{\psi}{\beta}\right)^{1/2} E_0^2 t_p. \tag{12}$$

If $t_p < t_c$ then we have a discharge that is initiated by the electron beam, i.e., after a short time a concentration $n_0 = \psi t_p$ is created in the gap, and then recombination decay of the plasma occurs according to the relation

$$n(t) = n_0 (1 + \beta n_0 t)^{-1}. \tag{13}$$

Here the specific energy deposited in the gas is defined as

$$w = \frac{e\mu E_0^2}{\beta} \ln \frac{n_0}{n_{cr}}, \tag{14}$$

where n_{cr} is a critical electron concentration defined in the following way: when the concentration in the cathode layer reaches n_{cr} the electric field is practically no longer distorted by the space charge, secondary processes no longer occur, and the discharge current ceases.[69] A non-self-sustaining discharge was first obtained with an initiating electron beam.[9,62] The current density in this case, according to (10) and (11), is $j = e\psi t_p \mu E_0$. Since $\mu \propto p^{-1}$ and $\psi \propto p$, j is independent of the gas pressure and proportional to the electric field. These conclusions are confirmed by experiment[62] (Fig. 8). From (11) and (12) it follows that in a non-self-sustaining discharge that is maintained by an electron beam the current density j is proportional to $j_p^{1/2}$ and the specific energy w is proportional to E_0^2. These same dependences are found experimentally (Fig. 9).[66]

Discharge ignition with external gas ionization by an electron beam is usually accomplished by injecting the beam into the gap through the cathode. At high gas pressures or large interelectrode spacings the electron energy may not be sufficient to produce uniform ionization throughout the width of the gap. It has been noted[62] that at low electron energies a volume discharge burns unstably and goes over into a spark discharge. Therefore, producing a uniform $\psi(x)$ is of particular concern in experiments with large interelectrode gaps.[73,76–79] The rate of gas ionization is given by the relation

$$\psi(x) = \frac{j_p D(x)}{e\bar{\varepsilon}}, \tag{15}$$

where $D(x)$ is the distribution of the energy loss per electron by the fast electrons in the gas, j_p is the beam current density, and $\bar{\varepsilon}$ is the average energy of formation of an electron-ion pair. The distribution $D(x)$, taking into account the electric field, is calculated by the multistep method,[73,80] or the Monte-Carlo method.[77,78,81] Use of these calculations has made it possible to account for the features of the I-V characteristics of a discharge with nonuniform ionization.[73]

If $\psi(x)$ varies with distance into the gap, the distribution of the electric field $E(x)$ is governed by the relation

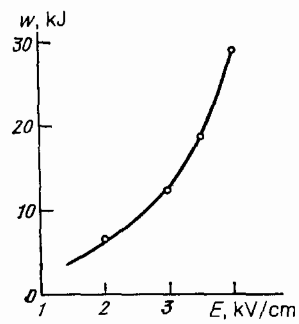

FIG. 9. Electric field dependence of the energy deposited in a 270 liter volume in a non-self-sustaining discharge.[66] Mixture CO_2:N_2:He = 1:1:3, beam current density $j_p = 0.4$ A/cm^2, $t_p = 1.2$ μsec.

$$E(x)=\left(\frac{\beta}{\psi(x)}\right)^{1/2}\frac{j}{e\mu}. \quad (16)$$

From (16) it follows that the electric field increases under conditions of weak ionization. The existence of a region of enhanced electric field leads to the formation and rapid development of a spark channel. Another reason for distortion of the field in the column may be the presence of thermalized beam electrons which form an uncompensated negative space charge in the gap. This causes an increase of the field in the region of the discharge column adjacent to the anode and also accelerates the contraction process. These contraction acceleration effects that stem from nonuniformities in $E(x)$ produced during the injection of the electrons into the gas have been called injection instabilities of the volume discharge.[79]

4. CONTRACTION OF A PULSED VOLUME DISCHARGE

4.1 General description of the contraction process

A characteristic feature of a volume discharge at high gas pressures is a limited burning time t_b, after which the discharge transforms into the channel form. This transition is accompanied by a redistribution of the current in the column and a localization of the current at the electrodes in the regions of cathode and anode spots. In the explanation of the contraction mechanism it was first widely held that the contraction is due to instabilities that arise in the discharge column.[71,82–84] A stability criterion in models proposed by various authors was that a certain threshold energy be released in the column. This approach correctly described the principal tendency, which is the decrease in t_b as the power supplied to the discharge is increased. However, it could not explain certain effects, such as the pressure dependence of the discharge burning time, the strong influence that the electric field in the column has on the discharge burning time, and other effects. Moreover, a number of models did not touch at all upon the question of how discharge channels form.

On the other hand experimental facts were obtained that showed that the transition to the channel stage is initiated by instabilities in the near-electrode regions. In the Townsend breakdown mechanism, during the volume discharge state, clumps in the near-cathode plasma caused by the formation of cathode spots have been observed,[85] as well as the transition to a channel discharge as a result of the propagation of the channel towards the electrodes.[86] The effect of cathode spot formation on the stability of a volume discharge and the formation of highly conducting channels as they propagate from the spots have been discussed for the case of self-sustaining volume discharges in Refs. 22 and 87, for discharges with ionization multiplication by an initiating electron beam in Ref. 88 and for non-self-sustaining discharges of 10^{-4} sec duration and steady-state discharges at atmospheric pressure in Refs. 89–91. In accordance with the concepts developed in Ref. 72, the transition from a volume discharge to a channel discharge takes place in two stages. First, disturbances are formed in the near-electrode regions (cathode and anode spots) and then highly conducting channels propagate from the spots. A detailed discussion of this contraction mechanism is given in Refs. 51 and 73.

4.2 Formation and behavior of cathode spots

Experimental investigations of volume discharges in a wide range of current density, 1–10^4 A/cm^2 and burning time, 10^{-4}–10^{-8} sec have shown that in most cases the transition to the channel stage is initiated with the formation of a cathode spot. An explosive emission mechanism for spot formation has been substantiated in which electric fields $E_c \approx 10^6$ V/cm and greater are present near the cathode.[79] There can be a variety of reasons why such fields are attained at different volume discharge current densities.[73] Let us first consider discharges with high current densities $j \gtrsim 100$ A/cm^2 and burning times $t_b \lesssim 10^{-6}$ sec. Here the critical field for microexplosions can be produced by the high concentration of positive ions in the cathode layer of the volume discharge.[79] For example, one can determine the field at the cathode in the region of anomalous current density for nitrogen and oxgyen from the approximations[51]:

$$\frac{E_c}{p}=1.1\cdot10^5\left(\frac{j}{p^2}\right)^{0.6}, \quad \frac{E_c}{p}=6.46\cdot10^4\left(\frac{j}{p^2}\right)^{0.47}, \quad (17)$$

where the dimensions of the quantities are: E_c, [V/cm] and j/p^2, [A/cm^2·torr^2]. It can be seen that for nitrogen at $p=1$ atm and normal current density $j_n/p^2=4\cdot10^{-4}$ A/cm^2·torr^2, the field at the cathode is $E_c=0.76\cdot10^6$ V/cm. With allowance for enhancement at the microscopic irregularities at the cathode surface, this field is sufficient to initiate explosive emission.[87]

This mechanism of spot formation is confirmed by the following experimental facts. At the instant of plasma flare formation near the cathode, emission lines of the atoms and ions of the cathode material are observed in the emission spectrum and microcraters are seen on the surface. The current density through the microcraters reaches $4\cdot10^8$ A/cm^2 (Ref. 51) and the craters themselves are attached to regions of enhanced electron emission.[92] The duration of the volume discharge is increased by eliminating the field emission regions by specially treating the cathode. By these means[92] a discharge was obtained in nitrogen with a current density $2\cdot10^3$ A/cm^2, a duration $t_b=300$ nsec, and a specific energy $w=15$ J/cm^2·atm. deposited in the gas.

In most experiments on volume discharges special measures to outgas the cathode surface are not taken and there is inevitably contamination, embedded insulating material, and oxide films on the cathode. Under these conditions, cathode spots form at low current densities when, according to (17), the field E_c certainly does not reach critical values. An important role is played by the charging of the outer surface of the contaminating films by the ion current[51,93] and as a result of this charging the field at the cathod grows to values that give rise to field emission, breakdown of the film, and the formation of a cathode spot. If it is assumed that the charge does not drain away from the film, then the time of formation of the cathode spot can be evaluated by the formula

$$t_p=\frac{E_{br}\varepsilon}{j}, \quad (18)$$

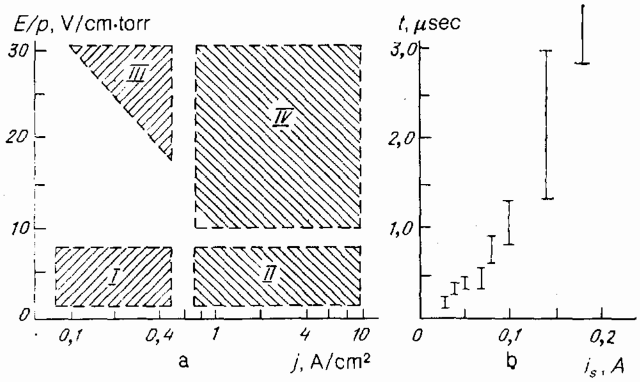

FIG. 10. (a) Regions I–IV of the different cathode spot burning modes and (b) dependence of stable spot operation on the current in the spot.[102]

where E_{br} is the breakdown voltage of the film, ε is the absolute dielectric constant, and j is the ion current density. This mechanism has been verified for the case of low-pressure glow discharges,[93] and for high-pressure discharges initiated by an electron beam.[51,73] In particular, relation (18) has been tested and it has been shown that a spot is initiated at a field $E_{br} = (2–4)\cdot10^6$ V/cm. A similar mechanism is also involved in initiating a spot on the anode,[51] since in the near-anode region the electron current dominates and insulating films can become charged by this current to the breakdown potential. A regular arrangement of spots on the cathode surface in a non-sustaining discharge has been observed.[94] It was found that this effect is caused by the development of an unstable cathode layer in the region of negative slope of the I-V characteristic. This interpretation made it possible to account for the regularity of the cathode spots.[73]

After the spot forms, part of the current of the volume discharge column contracts upon it and a highly conducting contracted spark channel propagates along the region of increased current density that is formed. The motion of the channel is due to the enhancement of the electric field at its apex and the increase in the specific power that is dissipated. One of the general experimentally observed features is that the electric field in the column has a greater effect on the propagation velocity of the channel than the current density has.[72] When E/p changes from (5–8) V/cm·torr to (30–35) V/cm·torr, the velocity at which the highly conducting channel grows from about the speed of sound (10^4 cm/sec[90,95–99] to 10^6–10^7 cm/sec.[88,100] In addition, the velocity increases with the gas pressure.[100] Some models describing the motion of the channel are discussed in Refs. 95–101.

We see then, that the conditions for the volume stage of the discharge are governed by the mechanism for cathode spot formation and subsequent channel development. By a generalization of the experimental results we can distinguish in the ranges of the discharge parameters E/p and j several regions for which specific modes of spot operation and channel development are characteristic. Such a distinction for a discharge initiated by an electron beam in nitrogen at $p = 100$ torr is shown in Fig. 10.[102]

For $E/p < 9$ V/cm·torr the transition from a volume discharge to a spark discharge is not observed. Region I of Fig. 10a corresponds to unstable spot burning, i.e., the discharge mode where the current through the spot is extinguished and then the spot is re-ignited. The dependence of the time of unstable spot burning on the current i_s through the spot is shown in Fig. 10b. As j increases, i_s also increases. When j is in region II the spot burns stably. In this region also, the spots are observed to propagate over the cathode surface and this leads to patterns in the form of branching channels on the cathode surface; photographs of this phenomenon are shown in Refs. 51 and 73. The increase of E/p leads to the propagation of a spark channel towards the anode. As the length of the channel increases, the current through it increases, and consequently the current through the cathode spot also increases. This leads to stable spot burning at low j but high E/p (region III).

In conclusion to this section, let us call attention to features of the contraction of the discharge in the active media of excimer lasers. Investigations of a discharge with external gas ionization with an electron beam[53] and of a self-sustaining discharge have shown that at low halogen concentrations (around 0.1% and less) the concluding phase of contraction — the growth of a highly-conducting channels from the spot — is not observed. At the cathode surface a large number of spots are formed, with diffuse channels attached to them. These formations overlap each other, giving a plasma column that is spatially uniform but on an area of the electrodes that is smaller than at first. The discharge is characterized by a low burning voltage and a conductivity that is maintained by the multistage ionization of the metastable atoms and molecules of the noble gases.[53,54] This phase has been called the high-current diffuse discharge. The duration of this discharge is 10^{-6} sec and greater. To prevent contraction in lasers based on noble gas halides with a self-sustaining discharge, schemes are usually used where it is possible to deposit the energy into the gas in the short time of 10^{-7} sec.[73] However, another alternative is not to permit the current density to increase as a result of contraction onto part of the cathode surface. This is accomplished by shaping the electrodes so as to obtain a uniform field in the discharge region and a distribution of spots over the entire area of the cathode.[51] Moreover, it is possible to control the current density by varying the inductance of the electrical circuit.[53] By these means it has been possible to obtain volume discharges with ionization multiplication that are suitable for pumping halide lasers with a discharge time of 10^{-6} sec,[53] and to obtain lasing in this regime.[104]

5. PULSED HIGH-PRESSURE CO_2 LASERS

5.1 Amplification coefficient and energy and spectral parameters of pulses during laser operation

The development of methods of igniting volume discharges has made it possible to pump high-pressure CO_2 lasers in the pulse length range 10^{-7} to 10^{-4} sec. It was shown already in the first experiments[63,69] that the principal features in the behavior of the kinetics of the processes at pressures up to tens of atmospheres remain the same as those at low pressures. Accordingly, all the merits of a discharge in

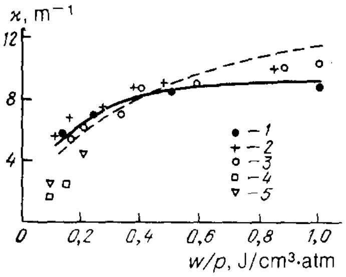

FIG. 11. Amplification as a function of specific energy for pumping a mixture of CO_2:N_2 = 1:2 by a discharge initiated by a 10 nsec beam of electrons.[109] p = : 1) 1 atm, 2) 2 atm, 3) 3 atm; curves 4,5: data of Ref. 69; dashed curve: calculation.

mixtures of CO_2:N_2:He as highly efficient laser active media in the infrared are preserved. In particular, pumping of the upper (00°1) laser level of the CO_2 molecule is accomplished by electron impact with transfer of the excitation from the first vibrational state ($v = 1$) of the nitrogen. Decay of the excited particles, aside from stimulated emission, occurs via vibrational relaxation during molecular collisions. A great deal of data on relaxation times[7,69,70] show that the increase in the reaction rate is totally determined by the gas pressure. For $p = 1$ atm, the lifetime of the upper lasing level is of the order 10^{-5} sec, which considerably exceeds the time of emptying the lower states of the symmetrical and deformation CO_2 vibrations, as well as the time of energy transfer from the N_2 to the CO_2.

Going to high gas pressures allows the solution of a number of fundamental problems in the physics and technology of gas lasers.

1. Because of the increased particle concentration the specific energy is increased by several orders of magnitude,[63,69] while the increased volume of the active medium makes it possible to obtain a total energy of 10^3–10^4 J.[66,105]

2. Average radiated power of the order of and greater than 10^4 W has been attained for lasers operating in cw and periodic pulse modes.[16,106]

3. Control of the lasing pulse shape and ultrashort pulses has been obtained.[7,16] Amplification modules with output energy greater than 10^3 J at a pulse length 10^{-9} sec have been made. On the basis of these devices, apparatus for inertial containment nuclear fusion at a total energy of 40 kJ have been developed and investigated (the "Antares" system).[107,108]

4. Continuous tuning of the lasing frequency over a wide range of the vibrational-rotational transitions of the *P* and *R* branches of other CO_2 molecule has been achieved.[69]

Let us consider the main features of CO_2 laser radiation. An important characteristic of the active medium is the amplification $\varkappa$. In general at the center of the lasing line, $\varkappa$ is proportional to the inversion density ΔN and inversely proportional to the width of the line. Typical conditions of laser excitation correspond to a specific energy $w \lesssim 0.3$ J/cm^3 input to the discharge, since at these levels there are still no deleterious effects due to heating of the active medium. On the basis of the pulse length t_p, or the specific energy dissipated in the gas, one can distinguish two limiting cases of excitation: $t_p \ll t_u$ and $t_p \gtrsim t_u$, where t_u is the lifetime of the upper laser level.

The results of measurements of the amplification at high pumping power are shown in Fig. 11.[109] A mixture CO_2:N_2 = 1:2 was excited in a discharge initiated by an electron beam, with $t_p = 2\cdot 10^{-7}$ sec. The measurements were carried out by the calibrated loss method in a laser with an active medium of 8 cm length. Under these conditions a high amplification is obtained and it can be seen that $\varkappa$ is determined by the specific energy deposition w/p. When $w/p \leqslant 0.4$ J/cm^2·atm, the inversion population ΔN is proportional to w/p, which also brings about a linear increase in $\varkappa$. At higher values of w/p the amplification grows less rapidly. The effect of the overlap of the vibrational-rotational spectral lines, which, for $p \geqslant 8$ atm causes an increase in $\varkappa$,[110,111] is not observed in these measurements.

For the other limiting case $t_p > t_u$ the results of measurements of the amplification by the method of probing with a weak signal are shown in Fig. 12.[112] Here a non-self-sustaining discharge of duration 10^{-4} sec, with specific power dissipation 2 kW/cm^2, is maintained by an electron beam ($j_p = 100\,\mu$A/cm^2). In contrast to the conditions shown in Fig. 11, the upper laser level is able to decay in the time t_p and consequently energy from the vibrationally excited nitrogen molecule can be pumped into it. This is the process that determines the dependence $\varkappa(t)$. The maximum amplification is attained after a time $([N_2] + [CO_2]) \times t_u / [CO_2]$, where $[N_2]$ and $[CO_2]$ are the concentrations of nitrogens and carbon dioxide in the gas), while the falloff of $\varkappa(t)$ is due to overheating of the active medium.

These modes of pumping that we have considered also result in considerably different laser pulse shapes. At low power the laser pulse shape duplicates the shape of the pumping pulse.[113] When the energy is pumped into the medium at high rates characteristic, for instance, for lasers with a self-sustaining discharge, a situation occurs in which the population inversion excess above the threshold comes about at a considerably greater rate than that at which the radiation field in the resonator is established. In this case one first observes a sharp peak of 10^{-7} sec duration in the radiation pulse that is associated with the emptying of the 00°1 state, and then a more prolonged pulse at a low power level. The shape of the tail of the radiation pulse is determined by the transfer of energy from the N_2 to the CO_2, by the decay of

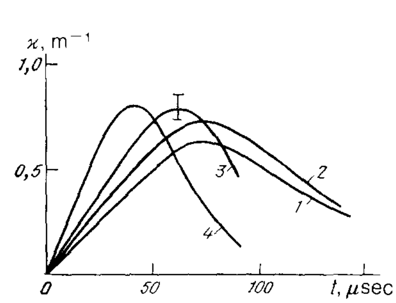

FIG. 12. Amplification as a function of time in quasistationary pumping mode for mixtures of CO_2:N_2:He in proportions: 1) 1:8:1; 2) 1:6:3; 3) 1:4:5; 4) 2:5:3; $p = 1$ atm, pumping power 2 kW/cm^3.

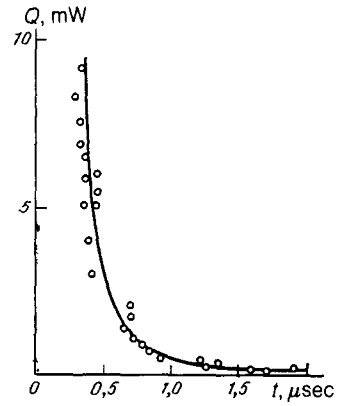

FIG. 13. Delay time for the first emission peak for various output emission power Q and, correspondingly, for various pumping power for a CO_2 laser with a self-sustaining discharge.[114] Curve: calculation; points: experimental data.

the lower level as well as the times required to establish higher order transverse modes in the resonator. Theoretical models for the CO_2 laser that have been developed allow us to predict this form of the laser pulse[7] and calculate the delay of the light pulse relative to the pumping current (Fig. 13).[114]

The output energy of a CO_2 laser is determined mainly by the pumping conditions, i.e., the ratio E/p during the burning stage of the discharge, by the power supplied to it, the composition of the mixture, etc. Under various pumping conditions the efficiency that is obtained, in terms of the output radiation energy relative to the energy fed into the volume, is in the range 10–30%. The most efficient lasers are those with a non-self-sustaining discharge and a pumping time $t_p \geqslant 10^{-6}$ sec. An example of efficiency optimization for a laser based on a non-self-sustaining discharge ($t_p = 10^{-6}$ sec) in a CO_2:N_2:He = 1:1:3 mixture and an active volume $10 \times 10 \times 100$ cm^3 is shown in Fig. 14.[115]

One of the characteristics of the laser transitions in the CO_2 molecule is that the conditions of self-excitation during lasing can be satisfied on a large number of transitions on the P and R branches. However, in the lasing spectrum at gas pressures $p \leqslant 5$ atm, a limited number of lines is usually seen.[116] The relative intensities of the typical lasing lines change with the external conditions (Fig. 15).[116] Calculations of the amplification spectra at high pressure taking into

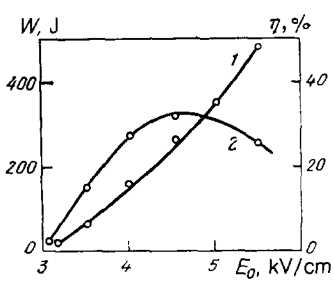

FIG. 14. Emitted energy and efficiency of a CO_2 laser with a non-self-sustaining discharge as a function of electric field.[115] Volume of active medium, 100 l, mixture CO_2:N_2:He = 1:1:3, $p = 1$ atm, $t_p = 10^{-6}$ sec.

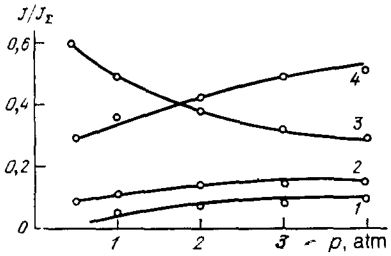

FIG. 15. Ratio of emission intensity J in the 00°1–10°0 transitions of the P branch to the total intensity J_Σ as a function of gas pressure[116] in a mixture CO_2:N_2 = 1:2. 1) P_{16}; 2) P_{18}; 3) P_{22}; 4) P_{20}.

account the overlap of the vibrational-rotational lines and deviation from equilibrium in the rotational degrees of freedom have been carried out in Refs. 69, 117, and 118. It is shown that lasing occurs on a number of lines, and the intense emission of the first peak leads to a redistribution of the populations of the rotational levels and correspondingly to a redistribution of the emission power in the individual transitions.

5.2 Laser emission tuning

It has been shown in Refs. 119 and 120 that the overlap of the rotational lines of the CO_2 molecule at high pressures makes possible continuous tuning of the lasing frequency. For this purpose it is necessary to use a selective resonator with a resolution higher than the frequency difference between the centers of the neighboring lines. Moreover, the pumping conditions must ensure that the amplification in the valley between the line is above the threshold level. In other words, one must select a sufficiently high parameter $\varkappa l$, where l is the length of the active region. Tuning of the CO_2 laser has been demonstrated experimentally.[120–124] Broadening of the tuning range was generally achieved by improved the resolution of the wavelength-selecting elements: diffraction gratings[120–122] and Fabry-Perot interferometers.[123,124] A tuning range up to 70 cm^{-1} was attained.

The choice of pumping conditions that provide the necessary value of $\varkappa l$ and increase of the aperture of the active volume are of great importance in solving the problems of increasing the emission energy of tunable lasers and broadening the tuning range. These problems were solved in Ref. 125. An active medium 100 cm long was formed by a non-self-sustaining discharge maintained by an electron beam with a current density 2 A/cm^2 and a pulse width $t_p = 1.2$ μsec. The resonator aperture was defined by a diaphragm 0.6 cm in diameter, an arrangement providing single-mode generation. The wavelength selecting element was a diffraction grating. It was shown that there is a limit to which the amplification can be increased, because the laser goes over into a self-excitation mode. Increasing the aperture entails difficulties because the single-mode condition of lasing is destroyed and transverse oscillations develop. The transverse modes are suppressed by putting special diaphragms in the resonator. In sum, the choice of pumping conditions has made it possible to created a laser with an emission energy of several joules that is tunable over the entire tuning range of 108

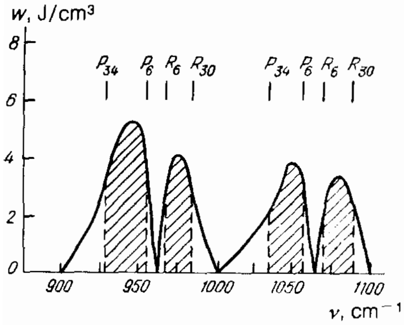

FIG. 16. Region of continuous tuning (crosshatching) in the laser transitions of the CO_2 molecule.[125]

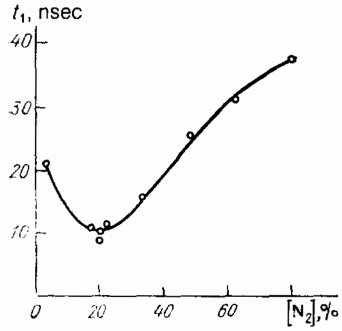

FIG. 17. Half-width of lasing pulse in a CO_2:N_2 mixture as a function of percent concentration of N_2.[130] Laser active region length 25 cm, $p = 3$ atm.

cm^{-1} (Fig. 16).[125]

5.3 Producing short emission pulses

One of the widely used ways of obtaining pulses 10^{-9} sec and shorter involves the phenomenon of mode locking.[7] Active,[126,127] passive,[128] and self-mode locking have been used. However, the trend to decreasing the CO_2 laser pulse width by increasing the pressure has stimulated investigations into obtaining short pulses in the free lasing regime. The general principles, developed in Ref. 117, of obtaining such pulses are these: it is necessary to decrease the length of the resonator and increase the pumping power while maintaining the product $\varkappa l_r$ constant. In addition, one should try to eliminate the second emission peak that is typical of high power, for instance by a choice of the proportion of CO_2:N_2 in the gas mixture. Experiments in shortening the pulses by this means have been carried out.[130,131] An active medium of volume $3.5\times4\times25$ cm^3 was created in a discharge initiated by a beam of current density 60 A/cm^2 and a 20-nsec pulse width.[130] The experiments were done with CO_2 and mixtures of CO_2:N_2:He. Increasing the pressure in pure CO_2 brought about a decrease in the lasing pulse width t_l. At $p = 4$ atm and a resonator length $l_r = 75$ cm, $t_l = 80$ nsec was obtained. Decreasing l_r to 40 cm reduced t_l by about a factor of three. Figure 17 (Ref. 130) shows the dependence of the pulse width on the per cent nitrogen concentration for a total pressure $p = 3$ atm. Here a width $t_l = 10$ nsec for the first peak was achieved. For these conditions single-frequency lasing on the transitions P_{16}–P_{24} is characteristic. In the work of Ref. 131 experiments were carried out for the purpose of obtaining short pulses in lasers with a self-sustaining discharge and $t_l = 15$ nsec was attained.

6. CONCLUSIONS

The use of pulsed volume discharges as the active medium for gas lasers has brought about a vigorous development in the physics and technology of lasers. Besides the results that we have presented here for the CO_2 systems, we can point out in addition many areas where successful development has been made possible by the solutions of the problems of creating active media at high pressure. In particular, new nontraditional mechanisms of laser pumping have been realized: pumping via recombination, charge transfer, the formation of complicated complexes, etc. A new type of laser has been discovered based on excimeric molecules of noble gas halides, where the necessary condition for pumping is a high gas pressure. At the present time this is the most intense source of stimulated emission in the ultraviolet. With the use of a volume discharge the reactions in chemical lasers are efficiently induced.

On the other hand, the requirements of gas laser physics stimulate investigations in very different areas, including the physics of gas discharges. Indeed, a new field "plasma for lasers" has arisen which encompasses study of the mechanism of the discharge conductance and devising means of calculating the I-V characteristic, determining the reasons for the transition from the volume stage of burning to the channel stage, addressing questions of plasma chemistry in a discharge as applied to various problems, problems associated with the theory of transport of fast electrons in dense gases, and with matching the power supply with the gas-discharge plasma, etc. Many important results have been obtained concerning these problems. There is no doubt that further investigations will broaden the concepts involved in volume discharges and will determine new possibilities for their practical application.

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