

## DYNAMIC PROCESSES IN A NITROGEN MOLECULAR LASER

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Physical processes in an active medium produced during development of the plasma of a powerful pulsed discharge in a neutral gas are considered. The low-pressure first-positive  $N_2$  band system of a laser is investigated quantitatively in experiment. It is shown that the appearance and growth of inversion population is due to a nonuniform electron excitation of working levels from the ground state that occurs following a rapid increase of the electron density. With the development of the plasma, the relative population of the working levels due to collisions between electrons and excited molecules attains the equilibrium distribution corresponding to the mean electron temperature. As a result the inversion population gradually disappears. The theoretical analysis is performed analytically for both rarefied and dense homogeneous active media for moderate and extremely rapid development of the plasma. Under optimal excitation conditions the dynamics of the active medium is such that the maximal inversion population is independent of the rate of electron density build-up and is proportional to the gas pressure. In the case of extremely rapid development of the plasma in a high-pressure gas, the mean electron energy is initially maximal and subsequently falls off rather rapidly. Relative stabilization of the electron energy then occurs; physically this is related to the characteristic break in the plot of ionization coefficient as a function of electron energy. In this case the electron density increases in an approximately linear manner. Under optimal excitation conditions the generation pulse duration is much shorter than the time of flight of the molecules in inelastic intermolecular collisions at gas pressures up to several tens of atmospheres.

## INTRODUCTION

IN the physics of gas lasers there exists an extensive region characterized by a sharp contrast between our very weak understanding of the observed phenomena and a wealth of experimental results. The empirical data on the existence of inverted population usually remain in this case without interpretation. This situation applies to lasers in which the active medium is a nonstationary nonequilibrium plasma produced on the front of a powerful pulse discharge in a gas<sup>[1-3]</sup>. If the experimental conditions are suitably chosen, generation on atomic, molecular, or ionic transitions can be produced with practically any substance in which a pulse discharge can be excited. Generation has been observed in one-third of all the elements of the periodic system.

The experimental results have received, at best, only a qualitative interpretation and have been considered quantitatively very rarely<sup>[4,5]</sup>. The formation of inversion at the start of a strong-current discharge is usually connected with electronic excitation of the working levels from the ground state<sup>[2-4,6]</sup>. A direct analogy of this inversion-excitation mechanism is the method of electromagnetic pumping<sup>[7]</sup> in the pulsed operating regime as proposed for the microwave region of the spectrum.

The main physical processes occurring in a plasma are known. In addition, there exist methods suitable for numerical calculations. However, the data required in this case concerning the main characteristics of the atoms and molecules (the effective collision cross sections and the radiative-transition probabilities) are ap-

proximate and insufficiently complete. It is therefore a rather complicated matter to obtain the physical picture of the processes with the accuracy that is required in this case by starting from a purely theoretical analysis. This is particularly true of the pulsed excitation regime. The theory of the nonstationary low-temperature plasma has not been sufficiently well developed. There is no information on the time variation of the electron density  $n_e$  and the average electron energy  $\epsilon$ . The great variety of working media excludes the possibility of a sufficiently rigorous general approach.

Under these conditions, to explain the occurring processes, it is useful to study a typical laser system. As a first approximation, it is quite reasonable to use simplified physical models chosen on the basis of the conditions in a real plasma.

This paper reports detailed investigations of the dynamic processes that lead to the occurrence and collapse of inversion in a molecular nitrogen laser operating on the first positive (1+) system of bands (electron transition  $B^3\Pi_g \rightarrow A^3\Sigma_u^+$ ,  $\lambda \cong 0.9 \mu$ ). There is no meeting of minds concerning the inversion mechanism in this laser. The inversion is attributed to inelastic intermolecular collisions in<sup>[1]</sup>, and to excitation of the working levels by electrons from the ground state in<sup>[8,9]</sup>. No quantitative verification of the proposed inversion mechanisms has been made. A comparison of theory with experiment is easiest to carry out for the amplification regime. To this end, measurement of the gain with a time sweep was undertaken. An approximate analytic theory of the laser is considered. The dynamics of the active medium at a high density of the working gas is discussed.

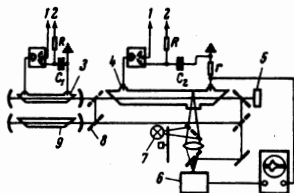


FIG. 1. Diagram of experimental setup: 1—discharge-gap ignition system, 2—rectifier 0 – +50 kV, 3—generator discharge tube, 4—amplifier tube, 5—thermocouple pile with galvanometer, 6—monochromator with diffraction grating and photomultiplier, 7—band lamp with mechanical modulator, 8—interchangeable flat mirrors, 9—tube of neon-helium laser.  $R = 500 \text{ k}\Omega$ ,  $C_1 = 0.01 \text{ }\mu\text{F}$ ,  $C_2 = 0.12 \text{ }\mu\text{F}$ ,  $r = 0.01 \text{ }\Omega$ .

## EXPERIMENTAL PROCEDURE AND MAIN RESULTS

The time dependence of the gain on individual rotational lines of the bands (1, 0) and (2, 1) was measured by shifting the generator light pulse relative to the amplifier (see the circuit in Fig. 1). The gain-measurement accuracy,  $\sim 10\%$ , was limited mainly by the stability of the generated-pulse amplitude, and the time resolution,  $\sim 0.2 \text{ }\mu\text{sec}$ , was determined by the duration of this pulse. The diameters and lengths of the tubes (in centimeters) were 0.4 and 80 for the generator and 1.5 and 200 for the amplifier. The population of the vibrational levels was determined from the relative and absolute intensities of the integral radiation of the individual bands. The latter was measured accurate to  $\sim 50\%$  with an oscilloscope, by comparison with the modulated radiation of the calibrated band lamp. The spontaneous emission was extracted through a side window sealed into the tube at a distance of 50 cm from the cathode.

We used in the experiments a discharge tube with electrodes of thoriated tungsten, filled with specially purified nitrogen at a pressure 0.8 mm Hg. The excitation-pulse repetition frequency usually did not exceed 0.5 Hz. The employed discharge ensured sufficiently high stability and reproducibility of the results.

The electron density  $n_e \gtrsim 10^{14} \text{ cm}^{-3}$  was determined interferometrically using a neon-helium laser at  $\lambda = 0.63 \text{ }\mu$ , and also using the time-varying contour of the  $H_\beta$  line of atomic hydrogen ( $n_e \gtrsim 10^{15} \text{ cm}^{-3}$ ). In the latter case,  $\sim 5\%$  hydrogen was added to the nitrogen. In the concentration region  $n_e \sim 10^{15} \text{ cm}^{-3}$ , the results of both methods coincide with accuracy  $\sim 50\%$ .

The dynamics of the excitation of different bands of the 1 + system of  $N_2$  is on the whole analogous. Figures 2–5 show some experimental results for the (1, 0) band<sup>1)</sup>. The upper and lower states of these bands are respectively  $B^3\Pi_g v' = 1 (B_1)$  and  $A^3\Sigma_u^+ v'' = 0 (A_0)$ . These states, as well as the ground state  $X^1\Sigma_g^+ v = 0 (X_0)$ , are also designated by the indices 1, 2, and 0.

Up to approximately the instant of cessation of the inversion  $t_c$ , the current density  $j$  increases exponen-

<sup>1)</sup>The experimental conditions for Figs. 2 and 3 are as follows:  $C = 0.12 \text{ }\mu\text{F}$ ,  $V = 13.5 \text{ kV}$ ,  $p = 0.8 \text{ mm Hg}$ , discharge length along the tube axis 2.0 m, distance between electrodes 2.3 m,  $E/p = 74 \text{ V/cm}\cdot\text{mm Hg}$ , tube diameter 1.5 cm, excitation pulse repetition frequency  $\sim 0.5 \text{ Hz}$ . For Fig. 5 the conditions are the same and the voltage is varied; for Fig. 4,  $C = 0.03 \text{ }\mu\text{F}$  and the total transmission of the resonator mirrors is  $\sim 2\%$ .

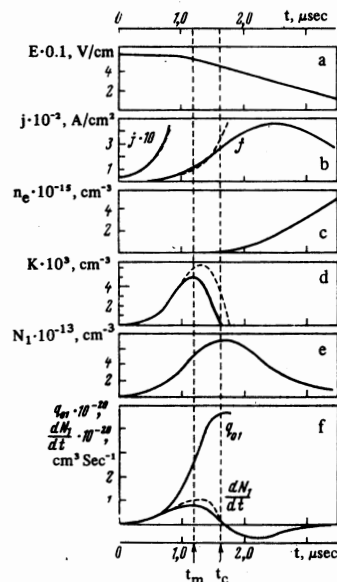


FIG. 2. Dynamics of excitation of inversion on the vibrational transition  $v' = 1 \rightarrow v'' = 0$  of the 1+ system of the  $N_2$  bands. a—electric field

$$E(t) = \frac{V_0}{d} - \int_0^t \frac{I(t)}{Cd} dt - \frac{L}{d} \frac{dI}{dt}$$

( $V_0$ —voltage on tube at the instant  $t = 0$ ,  $d$ —length of discharge gap,  $I$ —current,  $L$ —self-inductance of the circuit  $2.6 \text{ }\mu\text{H}$ ); b—density of discharge current; c—electron density, measured by interferometry; d—gain on the  $Q_{19}$  line of the (1, 0) band; e—absolute population of the vibrational level  $v' = 1$  of the B-state; f—rate of population of the  $B_1$  level by electrons from the ground state  $q_{01} = N_0 \langle \sigma_{01} v \rangle_t \cdot n_e(t)$  (see Footnote 2) and rate of change of the population of the  $B_1$  state  $dN_1/dt$ . Solid lines—results of experiment, dashed—calculation. Curves a and f were plotted by using curves b and e, respectively.

tially with time (Fig. 2b), the argument of the exponential being  $\sim 3 \times 10^6 \text{ sec}^{-1}$ . The characteristic current growth time is  $\sim 3 \times 10^{-7} \text{ sec}$ . The degree of ionization of the plasma at the instant  $t_c$  does not exceed 1%. During the initial section of the discharge, the population of the  $B_1$  state also increases exponentially (Fig. 2e) and linearly with the current (Fig. 3). The rate of excitation of the  $B_1$  state by electrons from the ground state  $q_{01} = N_0 \langle \sigma_{01} v \rangle n_e$  is in this case approximately equal to the population growth rate  $dN_1/dt$  (Fig. 2f). With further development of the discharge,  $q_{01}$  continues to increase and reaches a maximum, whereas  $dN_1/dt$  begins to decrease. The maximum of the gain does not coincide in time with the maximum population of the upper working state (Figs. 2d, e).

When the voltage is increased, the generation pulse duration  $\Delta t_g$  and the instant  $t_M$  of maximum intensity decrease (Fig. 4), the gain plot exhibits saturation, and the growth of the generation power gradually gives way to a decrease (Fig. 6). The increase of the generation power occurs with simultaneous increase of the gas pressure and of the voltage.

The instant  $t = 0$  on Fig. 2 corresponds to the start of the strong-current phase of plasma development. Owing to the low electron density at  $t < 0$  and to the rapid plasma relaxation, the complicated dynamic processes during the preliminary stages<sup>[10]</sup>, which in

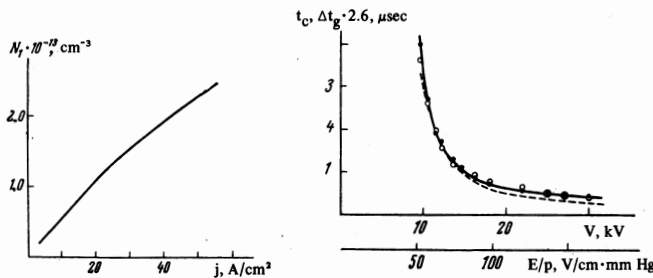


FIG. 3

FIG. 4

FIG. 3. Population of the  $B_1$  as a function of the current density.

FIG. 4. Dependence of the instant of maximum intensity  $t_M$  (●) and of the relative duration of the generation pulse  $\Delta t_g$  (○) on the voltage. Dashed line—calculation. The scale for the parameter  $E/p$  is given to make comparison with calculation convenient.

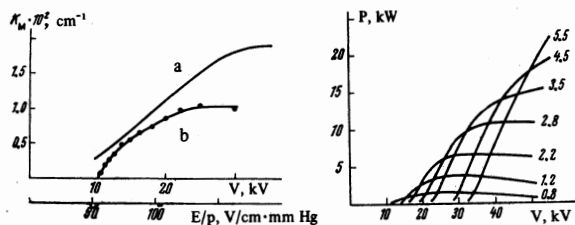


FIG. 5

FIG. 6

FIG. 5. Dependence of the maximum (with respect to time) of the gain at the  $Q_{1,9}$  line of the (1, 0) band on the voltage: a—calculation, b—measurements.

FIG. 6. Dependence of the generation peak power on the voltage at different gas pressures (indicated by the numbers at the curves in millimeters of mercury).  $C = 0.01 \mu\text{F}$ , length of discharge gap 1.3 m, tube diameter 3.0 cm, pulse repetition frequency 10 Hz, transmission of end mirror of resonator  $\sim 20\%$ .

many respects are still unclear, exert no noticeable influence on the population of the molecular states.

## DISCUSSION OF RESULTS

### A. Physical Models of a Pulse Plasma and of the Population Processes

The time variation of the current density  $j(t) = n_e v_d$  is determined in the general case by the time dependence of the electron density and the electron drift velocity. The electric field in the discharge can be regarded in this case as approximately uniform<sup>[10]</sup>. With the exception of a short initial time interval on the order of  $(\nu_{e0} + \nu_{ei})^{-1}$ , the form of the function  $v_d(t)$  is determined by the time variation of the electric field and by the summary effective collision frequency of the electrons with the molecules and the ions,  $\nu_{e0} + \nu_{ei}$ <sup>[11]</sup>. The time dependence of  $\nu_{e0} + \nu_{ei}$  is connected mainly with the rapid growth of the electron-ion collision frequency with increasing concentration of the charged particles. At a low degree of ionization (for a nitrogen plasma, according to estimates,  $\lesssim 4\%$ ), the influence of the Coulomb collisions is negligibly small and the drift velocity depends only on the field. As seen from Fig. 2a, the decrease of the electric field by the instant  $t_c$ , owing to the discharge of the

capacitance and the voltage drop on the self-inductance of the circuit, is negligible and the drift velocity can therefore be regarded as approximately constant. In the idealized case at  $L = 0$  and  $C = \infty$ , the field  $E$  and the drift velocity  $v_d$  are strictly constant in time.

At a constant drift velocity, the current density is  $j(t) = \text{const} \cdot n_e(t)$  and consequently, the characteristic growth times of the electron density and of the current coincide. Owing to the ambipolar diffusion on the walls, the lifetime of the electrons in the case of a diffusion-type initial electrons distribution over the discharge-tube cross section<sup>[12]</sup>,  $\tau_d = (r/2.4)^2/D_a = 2 \times 10^{-5}$  sec, is much larger than the characteristic growth time of the current  $\sim 3 \times 10^{-7}$  sec measured in the experiments ( $D_a$  is the coefficient of ambipolar diffusion,  $r$  is the tube radius, and the tube length is much larger than the radius). Neglecting the terms describing the electron loss due to diffusion and recombination ( $\tau_{rec} \gtrsim 2 \times 10^{-5}$  sec), in the electron-balance equation we obtain for the electron density  $n_e(t)$  and the current density  $j(t)$

$$n_e(t) = n_e^0 e^{\gamma t}, \quad (1)$$

$$j(t) = j_0 e^{\gamma t}, \quad j_0 = n_e^0 e v_d, \quad (2)$$

where  $\gamma = N_0 \langle \sigma_{oi} v \rangle = \alpha v_d$ ,  $N_0$  is the molecule concentration in the ground state,  $\langle \sigma_{oi} v \rangle$  is the product of the effective ionization cross section by the electron velocity, averaged over the velocity distribution of the electrons,  $\alpha$  is the Townsend ionization coefficient, and  $n_e^0$  and  $j_0$  are the initial electron density and current density. The generation is usually observed at  $\gamma \gtrsim 10^6 \text{ sec}^{-1}$ .

In practice, in the entire time interval  $t \lesssim t_c$ , in which inversion exists, the current growth can be described with satisfactory accuracy by formula (2) (see Fig. 2b). The argument of the exponential current growth ( $3.1 \times 10^6 \text{ sec}^{-1}$ ) is then approximately equal to the product of  $\alpha$  and  $v_d$ , which are known from<sup>[13,14]</sup> ( $\alpha v_d = 2.8 \times 10^6 \text{ sec}^{-1}$ ).

The time variation of the average electron energy  $\epsilon$  at a known  $n_e(t)$  dependence can be determined from the electron-energy balance equation. If the electric field  $E = E_0$  is "instantaneously" turned on,  $t \geq 0$ , a constant average electron energy  $\epsilon$  is established within the short time interval  $\sim 1/(\nu_{e0}\delta + \gamma)$  ( $\delta$ —average relative fraction of the energy lost by electrons by collisions with molecules)<sup>[11]</sup>. For  $E/p \lesssim 200 \text{ V/cm}\cdot\text{mm Hg}$ , only a negligible fraction  $\sim \gamma/\nu_{e0}\delta \ll 1$  of the energy input to the plasma is consumed in heating of the newly produced electrons. The average electron energy is in this case practically the same as in the case of a stationary plasma<sup>[15,16]</sup>, and is determined by the parameters  $E$ ,  $\nu_{e0}$  and  $\delta$ . Changes in the gas composition, which can influence the values of  $\nu_{e0}$  and  $\delta$  by the instant  $t_M$ , are small. Estimates based on the known effective cross sections for the ionization and excitation of the molecular states of  $N_2$ , with  $n_e(t)$  given in the form (1), show that the fraction of the ionized molecules and of the molecules in excited electronic states does not exceed 5%, and those in the vibrationally-excited states are  $\lesssim 10\%$ .

The analysis of the plasma dynamics, and subsequently also the interpretation of the main experimental

results, is greatly simplified in this case by choosing the experimental conditions. The cathode processes, allowance for which in the general form is quite complicated for the real state of the surface layer, have practically no influence on the dynamics of the strong-current phase of the discharge<sup>[10]</sup>. The plasma column is approximately homogeneous<sup>[10]</sup>, and the rate of plasma development is slow. A relatively complete set of data on the parameters of a low-temperature plasma in  $N_2$ , including those obtained theoretically on the basis of the kinetic equation<sup>[15]</sup>, also facilitates the analysis.

The experimentally obtained linear relation between the population of the upper working level and the current density (see Fig. 3) indicates electronic excitation of the levels from the ground state. The de-excitation of the levels by electrons in the initial instants of time is small compared with the electronic excitation from the ground state. It follows then from the equation for population, with  $n_e(t)$  and  $j(t)$  in the form (1) and (2), that the population of the upper working level  $B_1$ , in accordance with experiment, is proportional to the current density

$$N_1 = \frac{N_0 \langle \sigma_{01} v \rangle}{e v_d (\gamma + A)} j \quad (3)$$

( $A$  is the probability of radiative decay of the level  $B_1$ ). The slope of the experimental curve in Fig. 3 coincides with the proportionality coefficient in (3) accurate to a factor  $\sim 2.3$ . The agreement should be regarded as satisfactory, recognizing that the effective cross section of the electronic excitation of the  $B$  state is known with accuracy  $\sim 50\%$ <sup>[17]</sup>. Data on the Franck-Condon factors and on the radiative lifetimes of the  $B$  levels are given in<sup>[18,19]</sup>. The contribution of the radiative decay from the  $C^3\Pi_g$  state, the absolute population of which was measured in the experiments, to the population of the  $B_1$  state does not exceed 15%. Compared with the  $B_1$  level, electronic excitation of the lower working level  $A_0$  from the ground state is negligible, owing to the smallness of the corresponding Franck-Condon factor,  $f_{X_0A_0} = 6 \times 10^{-4}$ <sup>[18]</sup>.

The role of electron de-excitation processes increases with increasing population. The rate of growth of the population of the  $B_1$  level,  $dN_1/dt$ , decreases to zero and becomes subsequently negative, whereas the rate of the electronic excitation from the ground state  $q_{01}$  reaches a maximum<sup>2)</sup> (see Fig. 2f). Simultaneously, judging from the decrease of the gain, intensive population of the lower working state  $A_0$  begins.

The measured time dependence of  $dN_1/dt$  can be satisfactorily explained quantitatively within the framework of the following simplified population model: the level  $B_1$  is excited by electrons from the ground state, and de-excitation takes place via inelastic collisions

<sup>2)</sup>For  $t < t_c$  we have

$$q_{01} = N_0 \langle \sigma_{01} v \rangle n_e(t) \approx \frac{N_0 \langle \sigma_{01} v \rangle}{e v_d} j(t).$$

At  $t \sim t_c$ , the relative dependence of  $q_{01}$  was determined more accurately from the rate of excitation of the  $C^3\Pi_g$  state of  $N_2$  (the coefficients  $\langle \sigma_{X_0B_1} v \rangle$  and  $\langle \sigma_{X_0C} v \rangle$  depend on the electron energy in approximately the same manner). This took into account, by the same token, the decrease of  $q_{01}$  as a result of the decrease in the voltage.

of the electrons with the nitrogen molecules:  $N_2(B_1) + e \rightarrow N_2(A_V'') + e + \Delta\epsilon$  with a coefficient  $\langle \sigma_{B_1A} v \rangle = 6.8 \times 10^{-8} \text{ cm}^3 \text{ sec}^{-1}$  (dashed curve of Fig. 2f). The value of this coefficient, estimated on the basis of the Bethe approximation<sup>[20]</sup> with allowance for the vibrational structure of the  $B$  and  $A$  states, is  $2 \times 10^{-8} \text{ cm}^3 \text{ sec}^{-1}$ . (The oscillator strength of the transition is known from<sup>[21]</sup>.) The coefficients  $\langle \sigma v \rangle$  for ionization from the  $B_1$  level and for transition to the ground state are much smaller and amount, according to the estimates, to  $\sim 2.3 \times 10^{-9} \text{ cm}^3 \text{ sec}^{-1}$ , and  $2.5 \times 10^{-9} \text{ cm}^3 \text{ sec}^{-1}$ , respectively. The radiative lifetime  $\tau_B$  of the  $B$  state<sup>[19]</sup> is much longer than the time of existence of the inversion,  $\tau_B \gg t_c$ .

No resonant exchange of vibrational energy within the limits of the excited electronic state occurs within a time  $t < t_c$ , owing to the low frequency of the corresponding collisions ( $\lesssim 10^5 \text{ sec}^{-1}$ ). The considerable time needed to establish the vibrational relaxation<sup>[22]</sup> also excludes "mixing" of the populations of the vibrational levels as a result of collisions between the excited molecules and the molecules in the ground state.

## B. Interpretation of the Experimental Results

The dynamics of the inverted population is satisfactorily described on the basis of the mechanism of electronic excitation of the working levels from the ground state, with account taken of the quenching collisions of the electrons with the excited molecules for the idealized case of a homogeneous plasma with exponentially growing electron density and with constant electron temperature.

In the simplest case, when the electronic excitation of the upper working level 1 from the ground state 0 ( $0 \rightarrow 1$ ) and the quenching electronic collisions ( $1 \rightarrow 2$ ) are taken into account in the population equations,  $N_0 = \text{const}$ ,  $N_1^{t=0} = N_2^{t=0} = 0$ , the development of inversion proceeds as follows. At the initial instants of time the population of the upper working level increases linearly with electron concentration and exponentially in time  $N_1 = N_0 \langle \sigma_{01} v \rangle n_e^0 e^{\gamma t} / \gamma$ , while that of the lower state increases in proportion to the square of the electron density:

$$N_2 = \frac{N_0 \langle \sigma_{01} v \rangle n_e^0}{\gamma} \frac{\langle \sigma_{12} v \rangle n_e^0}{\gamma} e^{2\gamma t}.$$

Inversion sets in and increases exponentially for a certain time, owing to the fact that  $\langle \sigma_{12} v \rangle n_e^0 / \gamma \ll 1$ . Subsequently, owing to the more rapid decrease of the population  $N_2$  compared with  $N_1$ , the inversion reaches a maximum at an instant of time  $t_M$ , and then begins to decrease.

The dynamics of the active medium is in this case such that the inversion that is maximal in time

$$\Delta N_m = \frac{N_0 \langle \sigma_{01} v \rangle}{\langle \sigma_{12} v \rangle} \left( 1 - g_{12} \ln \frac{1 + g_{12}}{g_{12}} \right), \quad g_{12} = g_1 / g_2$$

( $g_1$  and  $g_2$  are the statistical weights of levels 1 and 2) does not depend on the growth rate of the electron concentration  $\gamma$ , and is proportional to the molecule concentration in the ground state. This is connected principally with the fact that with increasing  $\gamma$ , the value of  $t_M$  decreases approximately in inverse proportion to  $\gamma$ :

$$t_m = \frac{1}{\gamma} \ln \left[ \frac{\gamma}{\langle \sigma_{12} v \rangle n_e^0} \ln \frac{1 + g_{12}}{g_{12}} \right],$$

and  $n_e(t_M)$  increases in proportion to  $\gamma$ :

$$n_e(t_m) = n_e^0 e^{\gamma t_m} = \frac{\gamma}{\langle \sigma_{12} v \rangle} \ln \frac{1 + g_{12}}{g_{12}}.$$

Since the number of excited molecules at the instant  $t_M$

$$N_1(t_m) = N_0 \langle \sigma_{01} v \rangle \int_0^{t_m} n_e dt \approx N_0 \langle \sigma_{01} v \rangle n_e(t_m) t_m$$

is approximately proportional to the product  $n_e(t_M) t_M$ , it follows that  $N_1(t_M)$  is approximately equal, apart from logarithmic terms, to  $N_0 \langle \sigma_{01} v \rangle / \langle \sigma_{12} v \rangle$ , and is independent of  $\gamma$ . By the instant of time  $t_M$ , the inverse process  $2 \rightarrow 1$  comes into play in addition to the electronic collisions. This causes the relative population of the working levels to conform already at the instant  $t_C$  with the electronic temperature  $N_1/N_2 \approx g_{12} \exp(-\epsilon_{12}/\epsilon)$ , and the inversion stops ( $\epsilon_{12}$  is the energy of the working transition).

We present an expression for the inversion population  $\Delta N_{12} = N_1 - N_2 g_{12}$  with allowance, in the population equations, for the electronic excitation of the working levels from the ground state  $0 \rightarrow 1$  and  $0 \rightarrow 2$ , the ionization  $1 \rightarrow i$  and  $2 \rightarrow i$ , and the quenching collisions  $1 \rightarrow 0$  and  $1 \rightarrow 2$ <sup>3)</sup>:

$$\Delta N_{12} = N_0 g_{12} \left( \alpha_{02} + \frac{\alpha_{12}}{\alpha_1 - \alpha_{21}} \right) \left\{ \left[ 1 - \exp \left( -\frac{\alpha_1}{\gamma} (\eta - 1) \right) \right] \frac{\chi_1}{\alpha_1} - \left[ 1 - \exp \left( -\frac{\alpha_{21}}{\gamma} (\eta - 1) \right) \right] \frac{1}{\alpha_{21}} \right\}; \quad (4)$$

$$\chi_1 = \frac{\alpha_{01} (\alpha_1 - \alpha_{21} + g_{12} \alpha_{12})}{g_{12} [\alpha_{02} (\alpha_1 - \alpha_{21}) + \alpha_{12} \alpha_{01}]}, \quad \alpha_{01} = \langle \sigma_{01} v \rangle n_e^0,$$

$$\alpha_{i1} = \langle \sigma_{i1} v \rangle n_e^0, \dots, \alpha_1 = \alpha_{10} + \alpha_{12} + \alpha_{1i}, \quad \eta = e^{\gamma t}.$$

The average energy of the electrons, and together with it all the coefficients  $\langle \sigma v \rangle$  and  $\gamma$ , and also  $N_0$ , are assumed to be constant,  $n_e(t)$  takes the form (1), and  $N_1^{t=0} = N_2^{t=0} = 0$ . In the calculation of the coefficients  $\langle \sigma v \rangle$  for the states  $1+$  of the system of  $N_2$  bands, we used a Maxwellian electron-velocity distribution. The maximum of the function of the electronic excitation of the triplet upper working levels is reached at relatively low electron energies in the region where the deviations from the Maxwellian distributions are small. The dependence of the quantity  $\langle \sigma_{12} v \rangle$  on the average electron energy was calculated by starting from the Bethe approximation with the effective Gaunt factor<sup>[23]</sup>.

The maximum value of the inverted population

$$\Delta N_M = \frac{N_0 \alpha_{01}}{\alpha_1} \left\{ \frac{\alpha_{01} - g_{12} \alpha_{02}}{\alpha_{01}} - (1 - \chi_1^{-1}) \right\} \times \frac{g_{12}}{\alpha_{01} \alpha_{21}} \left[ \alpha_{02} (\alpha_1 - \alpha_{21}) + \alpha_{12} \alpha_{01} \right], \quad \xi = \frac{\alpha_{21}}{\alpha_1 - \alpha_{21}}, \quad (5)$$

is reached at the instant of time  $t_M$ :

$$t_M = \frac{1}{\gamma} \ln \left( \frac{\gamma}{\alpha_1 - \alpha_{21}} \ln \chi_1 \right). \quad (6)$$

For a Boltzmann distribution of the molecules over the vibrational states, with a gas temperature  $T_g$ , the

<sup>3)</sup>The processes  $1 \rightarrow i$ ,  $2 \rightarrow i$ ,  $1 \rightarrow 0$ , and  $0 \rightarrow 2$  are taken into account for greater generality. For a laser operating on the  $1+$  system of  $N_2$  bands these processes are insignificant under our experimental conditions.

inversion on the rotational transition  $B_v' j' \Lambda' \rightarrow A_v'' j'' \Lambda''$ , designated  $J' \rightarrow J''$  for short, is

$$\Delta N_{J', J''}(t) = \Delta N_{12}(t, g_{12}^*) g_n(2J' + 1) \frac{\exp(-\epsilon_{J', J''}/kT_g)}{Z_{v'}}, \quad (7)$$

where  $\Delta N_{12}(t, g_{12}^*)$  takes the form (4) with

$$g_{12}^* = g_e' \frac{Z_{v'} \exp(-\epsilon_{J', J''}/kT_g)}{Z_{v''} \exp(-\epsilon_{J', J''}/kT_g)}$$

in place of  $g_{12}$ . ( $g_n$  is the statistical weight of the level due to the spin of the nucleus,  $\epsilon_J$  is the energy of the rotational level,  $Z_v$  is the partition function of the vibrational state, and  $g_e' = 2$  is the electronic statistical weight of the upper working state). The gas temperature rise due to the elastic collisions of the electrons and of the ions with the molecules does not exceed  $\sim 50^\circ\text{C}$  at the instant  $t_M$ . The gas temperature  $T_g$ , determined by measuring the relative gain on the lines of the  $Q_3$  branch of the  $(1, 0)$  band, is  $\sim 360^\circ\text{K}$ .

The gain at the maximum of the Doppler-broadened rotational line is given by<sup>[24, 25]</sup>

$$K_{J', J''} = \frac{16(\pi^2 \ln 2)^{1/2}}{3hc} \frac{\nu}{\Delta \nu_d} \frac{S_e^{BA}}{g_e'} q_{v'v''} \frac{S_{J', J''}}{2J' + 1} \Delta N_{J', J''}. \quad (8)$$

In (8),  $\nu$  and  $\Delta \nu_d$  are the transition frequency and Doppler half-width of the line,  $S_e^{BA}$  is the strength of the electronic transition, and  $S_{J', J''}$  and  $q_{v'v''}$  are the Henle-London and the Franck-Condon factors.

For the amplification regime, the results of the calculations agree with the experimental data. The time dependence of the gain on the  $Q_{19}$  line of the  $(1, 0)$  band, calculated from formulas (8), (4), and (7), coincides with the experimentally measured curve accurate to 30–50% (see Fig. 2d). Agreement with experiment, especially for not too strong fields, is also observed in the voltage dependence of the time-maximum gain  $K_M$  on this line (see Fig. 5), and also in the instant  $t_M$  of the maximum inversion and in the relative duration of generation vs. the electric field (see Fig. 4).

Allowance for the population of the  $B_1$  level by electrons from the  $A$  state in the population equations introduces insignificant corrections in the results. The deterioration of the agreement between the theory and experiment that is observed in strong electric fields may be due to the increased role assumed by the inductive voltage drop in the case of fast plasma development.

The results of calculations of the saturated generation power  $P_{\text{sat}}^M$  disagree with the experimental data. The value of  $P_{\text{sat}}^M$  in the saturation regime  $N_1 \approx N_2 g_{12}$ <sup>[4]</sup> when account is taken of the electronic collisions  $0 \rightarrow 1$ ,  $1 \rightarrow 2$ , and  $2 \rightarrow 1$ , and also of the radiative decay  $1 \rightarrow 2$  with probability  $A$  in the population equations, is given by

$$P_{\text{sat}}^* = \frac{\hbar \nu}{1 + g_{12}} \frac{N_0 \gamma \alpha_{01}}{4 \alpha_{21} g_{12}} \frac{(1 - g_{12} A / \gamma)^2}{1 - \exp(-\epsilon_{12}/\epsilon)}, \quad (9)$$

and the energy of the generation pulse is

$$W_{\text{sat}} = \int_0^{t^*} P_{\text{sat}}(t) dt = 2 P_{\text{sat}}^* t^* / \gamma. \quad (10)$$

The maximum generation power in the saturation regime is proportional to the rate of excitation of the upper working level  $N_0 \langle \sigma_{01} v \rangle n_e(t_M)$ , where

$$t_M = \frac{1}{\gamma} \ln \left[ \frac{\gamma (1 - g_{12} A / \gamma)}{2 g_{12} \alpha_{21} (1 - \exp(-\epsilon_{12}/\epsilon))} \right].$$

The electron density is  $n_e(t_M) = n_e^0 e^{\gamma t} \approx \gamma$ , and therefore  $P_{\text{sat}}^M \sim N_0 \langle \sigma_{oi} v \rangle \gamma$  should theoretically increase rapidly with increasing voltage, owing to the factor  $\gamma$ . In fact, in the experiments (see Fig. 6) the generation power begins to decrease at sufficiently high voltage. In addition, the measured value of the peak power is smaller by one or two orders of magnitude than the calculated value.

These differences can be attributed to the finite development time of the photon cascade in the laser. The growth time of the photon cascade, from the level of spontaneous noise to the saturated photon density  $\sim 10^{16} \text{ cm}^{-3}$ , amounts to approximately  $10^{-7}$  sec, for the most intense rotational lines with maximum gain  $\sim 10^{-2} \text{ cm}^{-1}$ , as follows from the equation for the growth of the photon density in the oscillation mode. At  $E/p = 100 \text{ V/cm mm Hg}$ , this value is comparable with the time of existence of the inversion  $t_c$ . On the remaining aggregate of rotational lines making the principal contribution to the total calculated generation power, the gain is smaller, the photon cascade does not have time to develop, and no population inversion is realized. When the voltage is increased, the conditions for the development of a photon cascade become even worse: the growth of the gain slows down (see Fig. 5),  $t_{\text{cas}}$  becomes stabilized, and the time interval  $t_c$  decreases rapidly. The best conditions for the excitation of the laser can be ensured in excitation systems of the "traveling wave" type<sup>[26]</sup>, where the growth of the photon cascade is not limited by the time of existence of the inverted population.

The fraction  $\mu_B$  of the energy introduced per unit volume of the plasma owing to the excitation of the upper working state is ( $\mu_B$  is always smaller than unity for a spontaneous discharge):

$$\mu_B = \int_0^{t_c} N_0 n_e(t) \langle \sigma_{x,s} v \rangle \epsilon_B dt / \int_0^{t_c} n_e(t) E dt \approx \frac{N_0 \langle \sigma_{x,s} v \rangle}{e v_d E} \epsilon_B \quad (11)$$

(for  $t \lesssim t_c$  we have  $n_e \approx n_e^0 e^{\gamma t}$ ). For the B-state as a whole,  $\mu_B$  can reach  $\sim 35\%$ , and for the individual vibrational level  $B_{v'} = 1$  we have  $\mu_{B_1} = 5\%$ . The ratio of the generation pulse energy  $W_{\text{sat}}$  to the total introduced energy  $\epsilon(t_c)$  is

$$\frac{W_{\text{sat}}}{\epsilon(t_c)} = \frac{h\nu}{2(1+g_{12})} \frac{N_0 \langle \sigma_{oi} v \rangle}{e v_d E} \left(1 - g_{12} \frac{A}{\gamma}\right). \quad (12)$$

At optimal excitation conditions ( $g_{12}A/\gamma \ll 1$ ), as follows from (12), 1% of the expended electric energy goes over into coherent laser radiation.

The main results of the analysis carried out above for a laser operating on the 1+ system of  $N_2$  bands can be applied to any pulsed laser of similar type. As seen from (5) and (10), the inverted population and the generation pulse energy are proportional to the gas pressure, and the generation power (9) is proportional to the parameter  $\gamma = N_0 \langle \sigma_{oi} v \rangle$ . The quantity  $\langle \sigma_{oi} v \rangle$  under typical experimental conditions is usually smaller by one or two orders of magnitude than the optimal value (indicated by the arrow on Fig. 7). Raising the gas pressure and the electric field should lead to a rapid increase of the parameter  $\gamma$  and to a corresponding reduction of the discharge development time. Under these conditions, the dynamics of the active medium has certain distinguishing features.

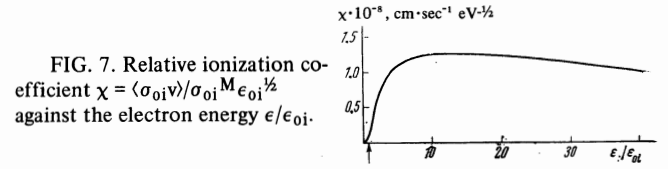


FIG. 7. Relative ionization coefficient  $\chi = \langle \sigma_{oi} v \rangle / \sigma_{oi}^M \epsilon_{oi}^{1/2}$  against the electron energy  $\epsilon/\epsilon_{oi}$ .

## EXCITATION OF DENSE ACTIVE MEDIUM OF A GAS LASER

A theoretical analysis of the dynamics of a weakly ionized plasma by numerically integrating the self-consistent system consisting of the kinetic equation and the circuit equation calls for rather lengthy calculations. The value of the results obtained in such a manner is furthermore certainly lowered by the approximate character of the initial data concerning the characteristics of the atoms and molecules. Within the framework of the simplest theory, the development of a weakly ionized plasma at a high current growth rate can be described approximately by a system of balance equations for the electron energy, the ionization, the conductivity, and the circuit. For an arbitrary function  $\gamma(E)$ , the solution of this system cannot be expressed in analytic form. At the initial instants of the development of a plasma in a neutral gas, the stepwise ionization and the electronic collisions can be disregarded, and the concentration of the atoms or molecules in the ground state can be regarded as constant.

For the  $\gamma(E)$  dependence we assume the following approximations:

$$\gamma(E) = \begin{cases} \gamma_m, & E_m \leq E \leq 3E_m \\ \gamma_m(E - E_+)/E_m, & E \leq E_m, \end{cases}$$

where  $E_+ \ll E_m$ ,  $E_m = (2/e) (\epsilon_{oi} \gamma_m m_e v_{\text{eff}})^{1/2}$ . The choice of this approximation is governed, on the one hand, by the available data on the drift velocity  $v_d$  and on the Townsend ionization coefficient  $\alpha$  ( $\gamma = \alpha v_d$ ), and on the other hand by the approximate  $\gamma(\epsilon)$  and  $\epsilon = f(E)$  dependences.

A comparatively complete set of data on the electron drift velocity and on the ionization coefficient  $\alpha$  is available only for  $E/p \lesssim 20 \text{ V/cm-mm Hg}$ <sup>[27]</sup>. In this region, the ionization rate  $\gamma$  for many gases is approximately proportional to  $eE/p$ . Above that ( $20-40 \lesssim E/p \lesssim 100-200 \text{ V/cm-mm Hg}$ ), judging from the available scanty data on the drift velocity, the growth of  $\gamma(E)$  becomes less steep, and for some gases ( $N_2, H_2, O_2, \dots$ ), the  $\gamma(E)$  curve becomes linear at  $E/p \approx 100-200 \text{ V/cm-mm Hg}$ . In view of the lack of data on  $v_d$ , it is impossible to trace the further course of  $\gamma(E)$ .

In choosing an approximation for  $\gamma(E)$  we can also start from the relation  $\gamma(\epsilon) = N_0 \langle \sigma_{oi} v \rangle = f(\epsilon)$ . The ionization coefficient  $\langle \sigma_{oi} v \rangle$  for different gases in the case of a Maxwellian electron velocity distribution,  $\langle \sigma_{oi} v \rangle \approx \sigma_{oi}^M \epsilon_{oi}^{1/2} \chi(\epsilon/\epsilon_{oi})$ , can be expressed approximately with the aid of the same function  $\chi(\epsilon/\epsilon_{oi})$  ( $\sigma_{oi}^M$ —maximum effective ionization cross section). This is possible because the relation  $\sigma_{oi}/\sigma_{oi}^M = f(\epsilon/\epsilon_{oi})$ , which enters in the expression for  $\chi$  changes relatively little on going from one gas to another (information on  $\sigma_{oi}$  can be found, for example, in<sup>[27]</sup>). The  $\chi(\epsilon/\epsilon_{oi})$  depend-

ence calculated on the basis of the data for H<sub>2</sub> is shown in Fig. 7. At low energies,  $\chi(\epsilon/\epsilon_{0i})$  decreases in proportion to  $\epsilon \exp(-\epsilon_{0i}/\epsilon)/\epsilon_{0i}$ . In accordance with Fig. 7, we can choose for  $\gamma(\epsilon)$  the following approximation:

$$\gamma(\epsilon) \approx \begin{cases} \gamma_m, & 4\epsilon_{0i} \leq \epsilon \leq 40\epsilon_{0i} \\ \gamma_m(\epsilon - \epsilon_+)/4\epsilon_{0i}, & \epsilon \leq 4\epsilon_{0i} \end{cases} \quad (13)$$

where  $\epsilon_* \ll 4\epsilon_{0i}$ . The approximate relation between  $\epsilon$  and  $E$  in the region of high energies  $4\epsilon_{0i} \lesssim \epsilon \lesssim 40\epsilon_{0i}$  ( $\gamma \equiv \gamma_m$ ) at  $E = \text{const}$  and  $\nu_{\text{eff}} = \nu_{e0} + \nu_{ei} = \text{const}$  can be found from the electron-energy balance equation:  $\epsilon \approx e^2 E^2 / \gamma_m m_e \nu_{\text{eff}}$ . At low electron energy  $\epsilon \lesssim \epsilon_{0i}$  the average energy  $\epsilon$  is in many cases approximately proportional to the field<sup>[27]</sup>. The approximation assumed above for  $\gamma(E)$  is obtained by assuming for simplicity  $\epsilon \propto E$  also in the intermediate energy interval  $\epsilon_{0i} \lesssim \epsilon \lesssim 4\epsilon_{0i}$ .

The time variation of the field and of the electron concentration is described by the following combination of the circuit and conductivity equations:

$$E\dot{n}_e + E(\dot{n}_e + \gamma n_e^*) = E_0 n_e^* \gamma_0, \quad (14)$$

where  $n_e^* = m_e \nu_{\text{eff}} d / e^2 S L \gamma_0$ ;  $d$  and  $S$  are the length and the cross section of the discharge gap,  $\gamma_0 = \gamma_{t=0}$ ,  $L$  is the self-inductance of the circuit,  $C = \infty$ , and the active resistance of the external circuit is  $r = 0$ . It is assumed that the initial electron concentration is  $n_e^0 \ll n_e^*$ . The field  $E$  is approximately constant up to an instant of time  $t^* = \ln(n_e^*/10 n_e^0) / \gamma_0$ , but then begins to decrease gradually. The average electron energy has time in this case to "follow" the variation of the field.

When  $E \sim E_m \gg E_*$  the ionization rate is approximately equal to  $\gamma(E) \approx \gamma_m E / E_m \approx \gamma_0 E / E_0$ , and the growth of the electron concentration in time is described by the following transcendental equation, which is obtained from (14) and from the ionization equation:

$$d\rho / d\tau + \ln \rho = \tau, \quad (15)$$

where  $\rho = n_e / n_e^*$ ,  $\tau = \gamma_0 t + \ln(n_e^0 / n_e^*)$ .

At the initial instants of time  $\tau \lesssim -2.3$ , the derivative in (15) can be neglected, the electron concentration increases exponentially,  $n_e(\tau) = n_e^* e^{\tau}$ , and the field is constant,  $E = E_0$ . Later on, at  $-2.3 \lesssim \tau \lesssim 14$ , the increase of the electron concentration and the decrease of the electric field are sufficiently well approximated by the expressions

$$n_e \approx 5 \cdot 10^{-4} (\tau + 6) n_e^*, \quad (16)$$

$$E \approx 4 E_0 / (\tau + 6). \quad (17)$$

The electric field is approximately equal to  $0.7E_0$  at  $\tau = 0$  ( $t = \ln(n_e^*/n_e^0) / \gamma_0$ ), and then decreases rapidly. The time interval  $t = \ln(n_e^*/n_e^0) / \gamma_0$ , during which a strong electric field and a high electron energy are maintained, decreases with increasing gas pressure, and  $E/N_0 = \text{const}$  is proportional to  $1/N_0$ . When the time  $\tau \approx 14$  is reached, the field  $E$  and the average electron energy decrease by an approximate factor of 5, and the electron concentration is  $\sim 80n_e^*$ . For the inductive development phase ( $\tau \gtrsim 14$ ,  $SLdj/dt \approx V_0$ ,  $n_e E \approx E_0 n_e^* \tau$ ,  $\gamma \approx \gamma_0(E - E_*) / E_0$ ) we obtain from the equation

$$d\rho / d\tau = \tau - E_+ \rho / E_0 \quad (18)$$

the following expression for the electron concentration:

$$n_e(\tau) \approx n_e^* \left[ \frac{E_0}{E_+} \tau - \left( 14 \frac{E_0}{E_+} - 80 \right) \exp \left\{ -\frac{E_+}{E_0} (\tau - 14) \right\} + \left( \frac{E_0}{E_+} \right)^2 \left( \exp \left\{ -\frac{E_+}{E_0} (\tau - 14) \right\} - 1 \right) \right]. \quad (19)$$

The electron concentration at  $\tau \gg E_0 / E_+$ , accurate to terms  $\sim \exp\{-E_+(\tau - 14)/E_0\}$ , increases linearly in time,  $n_e \approx n_e^* E_0 \tau / E_+$ , while the electric field and the average electron energy are constant:  $E \approx E_+$ ,  $\epsilon \approx \epsilon_*$ <sup>4)</sup>. Such behavior of  $n_e$  and  $\epsilon$  with time is physically connected with the fact that the  $\gamma(\epsilon)$  dependence has a sharp rise in the region of relatively low electron energies  $\sim \epsilon_*$ . The average electron energy established during the inductive phase should ensure the ionization rate necessary to maintain the linear growth of the current in time. The character of the connection between  $\gamma$  and  $\epsilon$  in the energy region  $\sim \epsilon_*$  is such that appreciable changes of the ionization rate correspond to a change of the electron energy in a narrow range about  $\epsilon_*$ . In the idealized case, when the approximation  $\gamma = \gamma_m(\epsilon - \epsilon_*) / 4\epsilon_{0i}$  is rigorously satisfied, the energy of the electrons in the inductive phase cannot drop below  $\epsilon_*$  at all. The relations  $\epsilon \approx \epsilon_*$ ,  $E \approx E_+$  and  $n_e \approx n_e^* E_0 \tau / E_+$  are not exact, since in the constant field and at  $\epsilon = \text{const}$  the electron concentration should grow exponentially and not linearly. The contradiction is eliminated by taking into account the terms  $\sim \exp\{-E_+(\tau - 14)/E_0\}$ .

For a strong initial field  $E_m < E_0 < 3E_m$ , the growth rate of the electron cascade is constant,  $\gamma \equiv \gamma_m$ , in the time interval  $\tau > \tau^*$ <sup>[28]</sup>, and then decreases in accordance with the decrease of the field as described by Eq. (15).

In the case of an arbitrary time variation of the excitation rate of the working levels, it is possible to obtain from the population equations the following expression for the generation power in the saturation regime:

$$P_n(t) = \frac{h\nu}{1 + g_{12}} N_0 (a_{01} - a_{02} g_{12}) \left\{ f(t) - \left[ \left( 1 - \exp \left( -\alpha \int_0^t f(t) dt \right) \right) + C \exp \left( -\alpha \int_0^t f(t) dt \right) \right] \frac{\beta + f(t)}{\alpha \cdot \delta} \right\}, \quad (20)$$

where

$$\alpha = \frac{g_{12} a_1 + a_2}{1 + g_{12}}, \quad \beta = \frac{g_{12} A}{a_{21} [\exp(\epsilon_{12}/\epsilon) - 1] + g_{12} (a_1 - a_2)},$$

$$\delta = \frac{a_{01} - g_{12} a_{02}}{g_{12} A (a_{01} + a_{02})}, \quad C = \frac{N^{l=0}}{N_0 (a_{01} + a_{02})} \alpha, \quad a_{01} = \langle \sigma_{0v} \rangle_{l=0} n_e^0,$$

$$a_{12} = \langle \sigma_{12v} \rangle_{l=0} n_e^0 = g_{12} \left( \exp \frac{\epsilon_{12}}{\epsilon} \right) a_{21},$$

$$a_1 = \sum_{k \neq 2} \langle \sigma_{1kv} \rangle_{l=0} n_e^0, \quad a_2 = \sum_{k \neq 1} \langle \sigma_{2kv} \rangle_{l=0} n_e^0;$$

$\sigma_{1k}$  and  $\sigma_{2k}$  are the effective cross sections for the de-excitation of molecules or atoms in the upper and lower working states by electrons,  $A$  is the probability of radiative decay on the working transition  $1 \rightarrow 2$ ,  $f(t) = \langle \sigma_{0kv} \rangle n_e(t) / n_e^0 \langle \sigma_{0kv} \rangle_{t=0}$  is a certain function of

<sup>4)</sup> This is confirmed in part by the results of numerical calculations performed for N<sub>2</sub> in [5].

time, and  $N_0 = \text{const}$ . It is assumed that the coefficients  $\langle \sigma \nu \rangle$  depend on the time in approximately the same manner. We consider the case when the lower working level is metastable, and the radiative decay of the upper working level to the ground state  $1 \rightarrow 0$  can be neglected. In Eqs. (20), no account was taken of the population and de-excitation of the levels as a result of the processes of radiative decay. This is valid if  $t_c \ll 1/A$ , where  $A$  is the characteristic radiative lifetime. If prior to the instant of the excitation pulse  $P = 0$ , then generation arises under the condition

$$\frac{dP_{\text{sat}}}{dt} = \left(1 - \alpha\beta \frac{f}{df/dt} - \alpha f \frac{f}{df/dt}\right) \exp\left(-\alpha \int_0^t f dt\right) - (1 - \alpha\beta\delta) > 0. \quad (21)$$

At the initial instants of time, the exponential term is approximately equal to unity and the condition is rewritten in the form

$$\delta > \frac{f}{df/dt} \left(1 + \frac{f}{\beta}\right). \quad (22)$$

For many laser systems, including lasers of the short-wave band, the influence of the quenching electronic collisions, at least at the initial instants of population, is small compared with the radiative decay, i.e., the term  $f/\beta$  in (22) can be neglected. To obtain stimulated emission in the saturation regime in such systems, the relative excitation rate of the active medium  $f^{-1}df/dt$  should be larger than the probability of radiative decay at the working transition:

$$\frac{1}{f} \frac{df}{dt} > g_{12} A_{12}. \quad (23)$$

The maximum generation power is determined from the condition  $dP_{\text{sat}}/dt = 0$ . For some of the simplest growing functions ( $e^{kt}$ ,  $t^n$ , ...) the generation power reaches a maximum at an instant of time  $t_M$  such that the exponential term in formulas (20) and (21) differs little from its value at  $t = 0$  in a wide range of parameters  $\alpha$ ,  $\beta$ ; and  $\delta$ . The generation power is then proportional to the gas pressure and, up to the instant of time  $t_M$ , is linearly connected with the rate of excitation of the active medium  $P_{\text{sat}} \approx h\nu N_0 (\alpha_{01} - g_{12}\alpha_{02}) \cdot f(t) / (1 + g_{12})$ .

The form of the function  $f(t)$  is determined by the character of the time variation of the electron energy and the electron concentration, and also by the dependence of the corresponding coefficients  $\langle \sigma \nu \rangle$  on the average electron energy. In the case of exponential plasma development, for  $t \lesssim \ln(n_e^*/10 n_e^0)/\gamma_0$ , the electron energy is  $\epsilon = \epsilon_0$ , and consequently the coefficients  $\langle \sigma \nu \rangle$  are constant, the electron concentration is  $n_e = n_e^0 \exp(\gamma_0 t)$ , and  $f = \exp(\gamma_0 t)$ . In the inductive phase of development ( $\epsilon \approx \epsilon_+$ ,  $\langle \sigma \nu \rangle \approx \text{const}$ ,  $n_e \propto t$ ), the rate of excitation of the levels is proportional to the time. Exceptions are optically-allowed transitions with excitation energy  $\epsilon_{01} \sim \epsilon_{0i}$  and  $\langle \sigma_{01\nu} \rangle \approx k \langle \sigma_{0i\nu} \rangle$ ,  $k = \text{const}$ , for which the rate of electronic excitation

$$N_0 \langle \sigma_{0i\nu} \rangle n_e(t) = N_0 \langle \sigma_{0i\nu} \rangle \langle \sigma_{0i\nu} \rangle n_e(t) / \langle \sigma_{0i\nu} \rangle \approx k n_e / dt$$

is constant in time when  $t \gtrsim E_0/\gamma_0 E_+$  and  $f = n_e^* E_0 / n_e^0 E_+$ .

With the exception of a short initial time interval  $\sim 10/N_0 \langle \sigma_{0i\nu} \rangle$ , the plasma development proceeds in the inductive regime. With increasing gas pressure, the duration of the initial section soon becomes

negligibly small. The maximum generation power  $P_{\text{sat}}^M$  in the saturation regime in the inductive phase of development is attained at the instant of time  $t_M$ :

$$P_{\text{sat}}^M \approx \frac{h\nu}{1 + g_{12}} N_0 (\alpha_{01} - \alpha_{02} g_{12}) \left( \frac{n_e^*}{n_e^0} \frac{E_0}{E_+} \gamma_0 \beta \delta \right)^{1/2}, \quad (24)$$

$$t_M \approx \left( \frac{n_e^0}{n_e^*} \frac{E_+}{E_0} \frac{\beta \delta}{\gamma_0} \right)^{1/2}, \quad (25)$$

the generation-pulse energy is then equal to

$$W_{\text{sat}} \approx P_{\text{sat}}^M t_M = \frac{h\nu}{1 + g_{12}} N_0 (\alpha_{01} - g_{12} \alpha_{02}) \beta \delta. \quad (26)$$

Expressions (24)–(26) follow from (20) at  $\langle \sigma \nu \rangle \equiv \langle \sigma \nu \rangle \epsilon_+ = \text{const}$ . The electron energy  $\epsilon_+$  in the inductive regime can be determined approximately by starting from the approximation (13) to the  $\chi(\epsilon/\epsilon_{0i})$  curve, see Fig. 7. We then obtain the value  $\sim 0.4\epsilon_{0i}$ . Somewhat more exact estimates of  $\epsilon_+$  can be obtained by starting from the real course of the  $\gamma(\epsilon)$  curve in the region of low energies, with allowance for the conductivity and circuit equations for the inductive regime ( $LdI/dt \gg E$ ,  $t \gg 14/\gamma_0$ ). These estimates yield  $\epsilon_+ \sim 0.22\text{--}0.15\epsilon_{0i}$ .

To illustrate expressions (24)–(26), the table lists certain numerical values for a laser operating on the infrared (1+ system of bands) and ultraviolet (2+ system of bands) transitions of molecular nitrogen. The products  $\sigma \nu$  for the upper and lower working levels were averaged over the Maxwellian distribution of the electron velocities. Information on the effective cross sections and on the Franck-Condon factors are contained in the literature references. Owing to the smallness of the Franck-Condon factor for the  $X_0 \rightarrow A_0$  transition, the corresponding coefficient  $\langle \sigma_{02\nu} \rangle$  is negligibly small. The effective cross sections  $\sigma_{ji}$  and  $\sigma_{2i}$  were estimated from the Thomson formula. To find the coefficients  $\langle \sigma_{10\nu} \rangle$  and  $\langle \sigma_{20\nu} \rangle$  we used the well-known relation for the direct and inverse processes. The value of the coefficient  $\langle \sigma_{12\nu} \rangle$  for the 1+ system of  $N_2$  bands was chosen starting from the data of the present investigation. For the 2+ system, the coefficient  $\langle \sigma_{12\nu} \rangle$  was chosen such as to obtain agreement with experiment<sup>[26]</sup> at low gas pressure. The calculations were performed at  $L = 10$  nH,  $d/S = 0.1$  cm<sup>-1</sup>,  $\epsilon_0 = 60$  eV,  $\epsilon_+ = 3.5$  eV,  $p = 1.5$  atm, and  $\gamma_0 = 4 \times 10^{12}$  sec<sup>-1</sup>. As seen from the Table, at a gas pressure  $\sim 1.5$  atm the per unit energy of the generation pulse, summed over the bands of the 1+ system, can reach 0.1 J/cm<sup>3</sup>, and for the (0, 0) band of the 2+ system we get 0.01 J/cm<sup>2</sup>.

The free-path time of the molecules in intermolecular collisions,  $\tau_n = 1/N_0 \langle \Sigma V \rangle$  ( $\Sigma$  is the effective cross section of the intermolecular collisions and  $V$  is the velocity of the molecules) becomes comparable with the generation duration at a molecule concentration  $N_0 \approx E_0 n_e^* \langle \sigma_{0i\nu} \rangle / E_+ n_e^0 \beta \delta \langle \Sigma V \rangle^2$ . For low-inductance excitation systems,  $n_e^* \approx 10^{12}$  cm<sup>-3</sup>, at  $E_0/E_+ \approx 20$ , the duration of generation in a laser system with  $\langle \sigma_{0i\nu} \rangle / \beta \delta n_e^0 \approx 10^{-15}$  cm<sup>6</sup> sec<sup>-2</sup> and  $\langle \Sigma V \rangle \approx 10^{-11}$  cm<sup>3</sup> sec<sup>-1</sup> is much shorter than the free-path time of the molecules in inelastic intermolecular collisions up to pressures  $\sim 30$  atm. Adiabatic collisions causing broadening of the spectral lines do not affect the level populations and therefore do not influence the generation power in the saturation regime.



$\langle\sigma v\rangle, \text{cm}^2\cdot\text{sec}^{-1}$							$A_{21}, \text{sec}^{-1}$	$P_{\text{sat}}^M, \text{MW/cm}^2$	$\Delta t_g, \text{nsec}$	$W_{\text{sat}}, \text{J/cm}^2$
$\langle\sigma_{01}v\rangle$	$\langle\sigma_{02}v\rangle$	$\langle\sigma_{11}v\rangle$	$\langle\sigma_{12}v\rangle$	$\langle\sigma_{21}v\rangle$	$\langle\sigma_{22}v\rangle$					
Working transition $B^3\Pi_g v' = 1 \rightarrow A^3\Sigma_u^+ v'' = 0$ (1 + system of $N_2$ bands, band (1, 0), $g_{12} = 2$ )										
$3.1\cdot 10^{-10}$ [17, 18]	$\lesssim 10^{-11}$ [18]	$2.3\cdot 10^{-9}$	$6\cdot 10^{-8}$	$2.5\cdot 10^{-9}$	$1.2\cdot 10^{-9}$	$\lesssim 10^{-10}$	$1.3\cdot 10^6$ [19]	15 (60)*	1.6	$0.025$ (0.1)*
Working transition $C^3\Pi_g v' = 0 \rightarrow B^3\Pi_g v'' = 0$ (2 + system of $N_2$ bands, band (0, 0), $g_{12} = 1$ )										
$3\cdot 10^{-10}$ [18, 29]	$1.2\cdot 10^{-10}$ [17, 18]	$2.4\cdot 10^{-8}$	$1.5\cdot 10^{-7}$	$2\cdot 10^{-9}$	$2.3\cdot 10^{-9}$	$2.5\cdot 10^{-10}$	$2\cdot 10^7$ [30]	13	0.6	$8\cdot 10^{-3}$

\*The parentheses contain the values summed over the bands (0, 0), (1, 0), (2, 1), and (3, 2).

## CONCLUSION

The measurements performed and the satisfactory quantitative agreement between theory and experiment confirm the mechanism of electronic excitation of inversion in a laser operating on the 1+ system of  $N_2$  bands from the ground state. The assumed weak population of the triplet states of  $N_2$  by electrons, on which the hypothesis of "stepwise" population is based<sup>[1, 31]</sup>, does not agree with data on the values of the corresponding excitation cross sections<sup>[17, 29]</sup>. The effect of the "smearing" of the radiation<sup>[31, 32]</sup>, to explain which this hypothesis was specifically proposed, can be connected naturally with the relatively large radiative lifetime of the B-state<sup>[19]</sup>.

The specific energy and the power of the coherent emission of pulsed gas lasers at sufficiently high working-gas pressure and at a corresponding value of the electric field can be comparable with the analogous parameters of solid-state lasers<sup>[33]</sup>.

Notice should be taken of certain difficulties that can arise when considerable volumes of the active medium are obtained. At a gas pressure above 0.01–0.1 atm, the discharge occurs under ordinary conditions in the form of a narrow spark, thereby violating the homogeneity of the active medium. This difficulty can apparently be avoided in part by preliminary homogeneous ionization of the gas<sup>[34, 35]</sup> or with the aid of the technique of needle-shaped cathodes with ohmic decoupling<sup>[36]</sup>.

With increasing gas pressure and increasing electric field, the rate of development of the plasma increases, and the duration of the generation is decreased (25). Violation of the homogeneity of the active medium is then possible as a result of the skin effect and pinch effect. With decreasing generation duration  $\Delta t_g$ , the electric energy from the remote sections of the energy-storage unit does not have time to reach the plasma gap, as a result of which the "effective" linear dimensions of the storage unit and the value of the energy input decrease in proportion to  $c\Delta t_g$ . For short stimulated-emission pulses ( $\lesssim 1$  nsec), in addition, the difficulties of the space-time matching in excitation systems of the "traveling wave" type increase<sup>[26]</sup>. To increase the generation duration it is expedient to use laser systems capable of effective excitation of the upper working levels at a low ionization rate. Suitable working levels may be chosen, for example, among the vibrational levels of the ground electronic states of molecules. Unlike optically allowed transitions, such levels can also be effectively excited in the inductive phase of plasma development, which is characterized by a relatively small value of the electron energy.

A pulse plasma offers prospects for obtaining stimulated emission in the short-wave region of the spectrum. The conditions of the excitation of generation improve with increasing relative rate of excitation of the working levels by the electrons,  $f^1 df/dt$  (see (23)). For a real plasma, this ratio is always finite. The minimum generation wavelength is then approximately equal to

$$\lambda_{\min} = \left( \frac{8\pi^2 e^2 f_{21}}{m_e c N_0 \langle\sigma_{01}v\rangle} \right)^{1/2}$$

( $f_{21}$  is the oscillator strength of the working transition). In excitation systems with the rapidly rising electron concentrations in the neutral gas and the  $\langle\sigma_{01}v\rangle \approx \text{const}$  that are characteristic of the breakdown of a neutral gas, the ratio  $f^1 df/dt = N_0 \langle\sigma_{01}v\rangle_m = \gamma_m$  is sufficiently large. In a laser system of similar type<sup>[3]</sup>, coherent radiation was recently obtained in the vacuum ultraviolet at a wavelength  $\lambda \approx 1600 \text{ \AA}$ <sup>[37]</sup>. However, it is difficult to excite generation with wavelengths shorter than  $\sim 1000\text{--}500 \text{ \AA}$  by this method, owing to the strong photoabsorption of the neutral gas. Further progress is apparently possible via pulsed excitation of inversion on transitions of multiply-charged ions during the process of rapid heating of the electrons in a fully ionized plasma.

The generation duration decreases in proportion to  $N_0^{1/2}$  in pulsed electronic excitation when the gas pressure is increased. This gives rise to the possibility of generating single ultrashort pulses of coherent radiation of limited power, with duration  $\sim 10^{-11}\text{--}10^{-12}$  sec, with sufficiently rigid synchronization relative to the triggering pulse. Spontaneous-emission light pulses of duration  $\sim 10^{-11}$  sec were observed in<sup>[38]</sup> in nitrogen at a pressure of several dozen atm.

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