

Investigation of a gasdynamic N₂O laser

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A theoretical and experimental investigation is carried out of the mechanism of operation and potentialities of a gasdynamic N₂O laser in which the working mixture escapes through a slit into vacuum. (Amplification occurs for the 00⁰1-10⁰0 transition, $\lambda \approx 10.9 \mu$.) The following mixtures are investigated: N₂O:N₂, N₂O:He, N₂O:N₂:He, N₂O:CO:He, and N₂O:N₂:H₂O. The kinetics of the physical processes is investigated. It is shown that, in contrast to a CO₂ laser, an important role is played in this case by nonresonant vibrational exchange which ensures additional pumping to the upper laser level. The dependences of the inverted population and of the gain on the major parameters are calculated. Experimental dependences of the gain on the pressure and temperature, on the distance from the slit, and on the nitrogen content in the N₂O:N₂ mixture, obtained by means of a shock tube, are in qualitative and, in some cases, in quantitative agreement with the theory. It is shown that the maximum inversion exceeds that obtained in a CO₂ laser and can be attained at considerably lower temperatures. These advantages of nitrous oxide over CO₂ are significant for practical purposes.

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1. INTRODUCTION

Interest in molecular gas lasers has not diminished in the last few years. Among these lasers, those based on vibrational transitions of the CO₂ molecules are sharply outstanding in efficiency, simplicity, and practical importance. High efficiencies were obtained in electric-discharge CO₂ lasers, and unprecedented powers in the quasicontinuous regime were obtained in gasdynamic lasers (GDL). For applied purposes, in addition to improving the characteristics of CO₂ lasers, great importance attaches to searches and investigations of new working media. At one time, definite interest was evidenced in the use of the N₂O molecule in lasers, since its system of vibrational levels is quite similar to the level system of CO₂.^[1] The very first investigations^[2-4], however, have shown that electric-discharge N₂O lasers deliver much less power than CO₂ lasers, owing to the appreciable dissociation of the N₂O in the discharge. Therefore practically no further attention was paid to the investigation of N₂O lasers. In the GDL, however, the effect of dissociation is not so significant and in principle one can expect results close to those obtained in CO₂ lasers (see^[5]). Indeed, recent experimental results on N₂O GDL, obtained with a shock tube, confirmed this statement. Thus, the inverted population in N₂O was experimentally investigated following adiabatic expansion of a heated N₂O:N₂:He mixture by the weak-signal amplification method^[6]. The result was a large value of the gain, $\sigma_{\max} = 1.15 \times 10^{-2} \text{ cm}^{-1}$. Generation from a GDL using an N₂O:N₂:He mixture was realized in practice for the first time by expansion through a nozzle^[7].

The present paper is devoted to a comprehensive theoretical and experimental investigation of the operating mechanism and of the possibilities of an N₂O laser. The analysis is carried out for the mixtures N₂O:N₂, and N₂O:N₂:He, N₂O:CO:He, N₂O:N₂:H₂O, which were heated by a reflected shock wave and escaped through a slot into vacuum. A theoretical analysis of the kinetics of the physical processes in the working mixtures was used to calculate the dependences of the inverted population of the vibrational levels 00⁰1-10⁰0 N₂O ($\lambda \approx 10.9$) and of the gain α on the parameters of the system. The results of a detailed experimental investigation are in

qualitative agreement, and in a number of cases also in quantitative agreement, with the calculation. This has made it possible not only to study the operating mechanism of the N₂O laser, but also to demonstrate, under definite conditions, its advantages over CO₂ lasers. This is very important from the applied point of view, and therefore the experimental data for the N₂O GDL are systematically compared not only with the calculated relations but also with the corresponding relations for CO₂ GDL.

2. KINETICS OF VIBRATIONAL PROCESSES. FUNDAMENTAL EQUATIONS

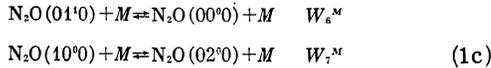
a) Principal Relaxation Channels for the N₂O Molecule

The structure of the vibrational levels of the N₂O molecule is similar in many respects to the structure of the CO₂ molecule (Fig. 1), but has also its own distinguishing features. Thus, although the energy defect between the 00⁰1 level of N₂O and the first vibrational level of N₂ is small and amounts to 107 cm⁻¹, it is larger than the distance between the CO₂ and N₂ levels (19 cm⁻¹). In addition, all the levels of the N₂O molecule lie somewhat lower than the corresponding CO₂ levels, so that less energy is needed to pump N₂O. It is also important that near the 00⁰1 state of N₂O there are more levels capable of exchanging energy with the 00⁰1 level.

These features of the N₂O molecules were taken into account in the comparison of the relaxation equations. In the scheme assumed by us, the principal processes that determine the balance of the vibrational energy in the N₂O:N₂:He are assumed to be the following:

$$N_2O(00^0 1) + M \rightleftharpoons \begin{cases} N_2O(11^1 0) + M & W_1^M \\ N_2O(12^0 0) + M & W_2^M \\ N_2O(20^0 0) + M & W_3^M; \\ N_2O(03^1 0) + M & W_4^M \\ N_2O(04^0 0) + M & W_5^M \end{cases} \quad (1a)$$

$$N_2O(00^0 0) + N_2(v=1) \rightleftharpoons \begin{cases} N_2O(00^0 1) + N_2(v=0) & W \\ N_2O(11^1 0) + N_2(v=0) & W_1 \\ N_2O(12^0 0) + N_2(v=0) & W_2; \\ N_2O(20^0 0) + N_2(v=0) & W_3 \\ N_2O(03^1 0) + N_2(v=0) & W_4 \\ N_2O(04^0 0) + N_2(v=0) & W_5 \end{cases} \quad (1b)$$



Here $M = \text{N}_2\text{O}$, N_2 , or He . On the right are shown the symbols for the probabilities of the corresponding processes (the rate constants of the vibrational relaxation).

b) Vibrational-Kinetics Equations

The general form of the vibrational-relaxation kinetics of a multicomponent mixture of harmonic oscillators, assuming the presence of total equilibrium within each individual mode, was obtained in [8]. Assuming for simplicity that $h\nu_1 = 2h\nu_2$ and that the vibrational temperatures of the symmetrical (T_1) and deformation (T_2) vibrations are the same, we obtain in analogy with [8, 9]

$$\begin{aligned} \frac{d\epsilon_1}{dx} &= \frac{N}{V} \left\{ W \left[e^{-\epsilon_1/kT} \epsilon_3 (1+\epsilon_1) - \epsilon_1 (1+\epsilon_3) \right] \right. \\ &+ W_{1,z_1} \left[e^{-\epsilon_1/2kT} \left(\frac{\epsilon_2}{2} \right)^3 (1+\epsilon_1) - \epsilon_1 \left(1 + \frac{\epsilon_2}{2} \right)^3 \right] \\ &+ W_{1,z_2} \left[e^{3\epsilon_2/kT} \left(\frac{\epsilon_2}{2} \right)^4 (1+\epsilon_1) - \epsilon_1 \left(1 + \frac{\epsilon_2}{2} \right)^4 \right] \left. \right\} \gamma_{\text{N}_2\text{O}}, \\ \frac{d\epsilon_2}{dx} &= \frac{N}{V} \left\{ -W \gamma_{\text{N}_2} \left[e^{-\epsilon_2/kT} \epsilon_3 (1+\epsilon_2) - \epsilon_2 (1+\epsilon_3) \right] \right. \\ &+ \left[e^{-\epsilon_2/kT} \left(\frac{\epsilon_1}{2} \right)^3 (1+\epsilon_2) - \epsilon_2 \left(1 + \frac{\epsilon_1}{2} \right)^3 \right] \sum_M W_{3,z_1}^M \gamma_M \\ &+ \left[e^{19\epsilon_1/kT} \left(\frac{\epsilon_1}{2} \right)^4 (1+\epsilon_2) - \epsilon_2 \left(1 + \frac{\epsilon_1}{2} \right)^4 \right] \sum_M W_{3,z_2}^M \gamma_M \left. \right\}, \\ \frac{d(\epsilon_2+2\epsilon_1)}{dx} &= \frac{N}{V} \left\{ -3W_{1,z_1} \gamma_{\text{N}_2} \left[e^{-\epsilon_2/kT} \left(\frac{\epsilon_1}{2} \right)^3 (1+\epsilon_1) - \epsilon_1 \left(1 + \frac{\epsilon_2}{2} \right)^3 \right] \right. \\ &- 3 \left[e^{-\epsilon_2/kT} \left(\frac{\epsilon_1}{2} \right)^3 (1+\epsilon_2) - \epsilon_2 \left(1 + \frac{\epsilon_1}{2} \right)^3 \right] \sum_M W_{3,z_1}^M \gamma_M \\ &- 4W_{1,z_2} \gamma_{\text{N}_2} \left[e^{3\epsilon_1/kT} \left(\frac{\epsilon_1}{2} \right)^4 (1+\epsilon_1) - \epsilon_1 \left(1 + \frac{\epsilon_2}{2} \right)^4 \right] \\ &- 4 \left[e^{19\epsilon_1/kT} \left(\frac{\epsilon_1}{2} \right)^4 (1+\epsilon_2) - \epsilon_2 \left(1 + \frac{\epsilon_1}{2} \right)^4 \right] \sum_M W_{3,z_2}^M \gamma_M \\ &\left. + (1-\beta) (\epsilon_{02} - \epsilon_2) \sum_M W_{02}^M \gamma_M \right\}, \end{aligned} \quad (2)$$

where

$$\epsilon_i = r_i \exp(-h\nu_i/kT) / [1 - \exp(-h\nu_i/kT)]$$

is the average reserve of the quanta of the i -th vibration of one molecule, r_i is the multiplicity of oscillator degeneracy ($r_1 = r_2 = r_4 = 1$; $r_3 = 2$), T_i is the vibrational temperature of the i -th oscillator, x is the coordinate along the expansion axis, $\epsilon_{02} = \epsilon_2(T)$, $\beta = \exp(-h\nu_2/kT)$, $V = V(x)$, $N = N(x)$ are the local velocity and density (cm^{-3}) of the gas, γ_M is the molar fraction of the M -th component, while the dimension of $[W]$ is cm^3/sec . Under these assumptions, $\epsilon_1 = (\epsilon_2/2)^2(1+\epsilon_2)^{-1}$, and the equations for ϵ_1 and ϵ_2 reduce to a single equation for ϵ_2 . In this case

$$\frac{d(\epsilon_2+2\epsilon_1)}{dx} = \frac{2+6\epsilon_2+3\epsilon_2^2}{2(1+\epsilon_2)^2} \frac{d\epsilon_2}{dx}.$$

To simplify the notation, we have introduced in (2) the symbols

$$\begin{aligned} W_{1,z_1} &= W_1 + \frac{W_1}{1+\epsilon_2}, & W_{1,z_2} &= W_2 + \frac{W_2}{1+\epsilon_2} + \frac{W_3}{(1+\epsilon_2)^2}, \\ W_{3,z_1}^M &= W_1^M + \frac{W_1^M}{1+\epsilon_2}, & W_{3,z_2}^M &= W_2^M + \frac{W_2^M}{1+\epsilon_2} + \frac{W_3^M}{(1+\epsilon_2)^2}. \end{aligned} \quad (3)$$

c) Transition Probabilities

The question of the choice of probabilities was discussed in detail in [10]. We dwell here only on the final expressions used by us in the calculations.

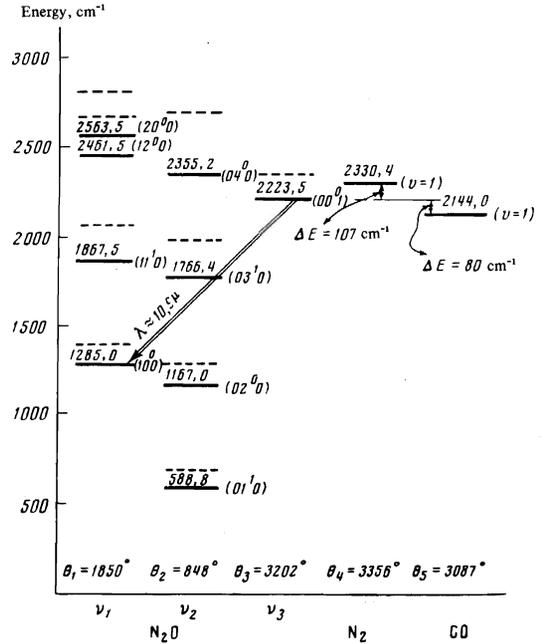


FIG. 1. Scheme of lower vibrational levels of the molecules N_2O , $\text{N}_2(v=1)$, and $\text{CO}(v=1)$. The dashed lines show the corresponding levels of the CO_2 molecule, and the arrow indicates the laser transition.

To determine the relaxation probability of the 00^0_1 level we used experimental data on the relaxation time τ_{exp} . In analogy with [11], it has been assumed that during the measurement of τ_{exp} the deformation and symmetrical vibrations are almost in equilibrium with the translational degrees of freedom. The equation for ϵ_3 in the case of the $\text{N}_2\text{O}:\text{He}$ mixture can then be represented in the form

$$\frac{d\epsilon_3}{dt} \approx \frac{\epsilon_{03} - \epsilon_3}{\tau_{\text{exp}}}, \quad (4)$$

where

$$\begin{aligned} \tau_{\text{exp}} &= \left[N \sum_M \gamma_M W_{\text{exp}}^M(00^0_1) \right]^{-1}, \\ W_{\text{exp}}^M(00^0_1) &\approx W_3^M (1-\beta)^{-3} f^M(T) \quad [10], \\ f^M(T) &= \left[\frac{W_1^M}{W_3^M} + \left(\frac{W_2^M}{W_3^M} + \frac{1}{1+\epsilon_2} \right) \frac{1}{1-\beta} \right] \frac{1}{1+\epsilon_2}. \end{aligned} \quad (5)$$

Expressions for the probability ratios in $f^M(T)$ were calculated by the procedure of [12] and are given in [10]. (The reliability of the calculation of the relative probabilities is higher than that of the absolute ones.) When nitrogen-containing mixtures were considered, account was taken of the channel for energy exchange between the levels 00^0_1 and $v=1$ of the N_2 molecule. The connection between $W_{\text{exp}}^M(00^0_1)$ and the probabilities that enter in (2) were determined in analogy with [13]:

$$\begin{aligned} W_{3,z_1}^M &= \frac{W_1^M}{W_3^M} \frac{(1-\beta)^3}{1+\epsilon_2} \frac{W_{\text{exp}}^M(00^0_1)}{f^M(T)}, \\ W_{3,z_2}^M &= \left(\frac{W_2^M}{W_3^M} + \frac{1}{1+\epsilon_2} \right) \frac{(1-\beta)^3}{1+\epsilon_2} \frac{W_{\text{exp}}^M(00^0_1)}{f^M(T)}. \end{aligned} \quad (6)$$

A similar procedure was used in the analysis of the processes of energy transfer to the levels ($v_1 v_2 l_0$) of the N_2O molecule in collisions with $\text{N}_2(v=1)$. It turned out that the calculated probabilities W_i [Eq. (1b)] are much smaller than the corresponding W_i^M [Eq. (1a)]. To simplify the calculations the processes of disintegration of $\text{N}_2(v=1)$ via the levels of N_2O (i.e., $W_{4\Sigma_1}$ and $W_{4\Sigma_2}$) were therefore excluded from consideration.

To solve the system (2) it is necessary to specify the temperature dependences of the exchange rate W and of the relaxation rates W_{exp}^M of the upper level (00^01) and W_6^M of the lower level (01^01). The rates of exchange and relaxation have been measured in a number of studies^[14-21]. The temperature dependences for W , $W_{\text{exp}}^{\text{N}_2\text{O}}$ (00^01) and $W_6^{\text{N}_2\text{O}}$ were taken respectively from^[14, 15, 16]. For the disintegration of the upper levels $W_{\text{exp}}^{\text{N}_2}$ (00^01), $W_{\text{exp}}^{\text{He}}$ (00^01), and the lower level $W_6^{\text{N}_2}$, W_6^{He} of the N_2O molecule in collisions with N_2 and He there are only room-temperature data. We therefore chose for them the same temperature dependences as for the corresponding processes in CO_2 (in turn, the temperature dependences for CO_2 were obtained on the basis of^[22]) and matched them to the experimentally measured values^[17-21] at $T = 300^\circ\text{K}$. This choice of temperature dependences for $W_{\text{exp}}^{\text{N}_2}$ (00^01), $W_{\text{exp}}^{\text{He}}$ (00^01) and $W_6^{\text{N}_2}$, W_6^{He} is valid since, first, the arrangement of the N_2O levels is analogous to that of the corresponding CO_2 levels (Fig. 1), and second, the temperature dependences of $W_{\text{exp}}^{\text{N}_2}$ (00^01) and $W_6^{\text{N}_2\text{O}}$ given in^[15, 20] for N_2O are approximately similar in character to the corresponding dependences for CO_2 . The probabilities assumed by us are

$$\begin{aligned}
 W &= 1.1 \cdot 10^{-16} T [3 + (T - 600)^2 \cdot 1.33 \cdot 10^{-5}] \exp(154/T), \\
 W_{\text{exp}}^{\text{N}_2\text{O}}(00^01) &= 1.1 \cdot 10^{-16} T \cdot 10^{3.28 - 23.6T^{-0.5}}, \\
 W_{\text{exp}}^{\text{N}_2}(00^01) &= 7.5 \cdot 10^{-10} \exp(-75.5T^{-0.5}), \\
 W_{\text{exp}}^{\text{He}}(00^01) &= 2.9 \cdot 10^{-12} T^h \exp(-58.5T^{-0.5}), \\
 W_6^{\text{N}_2\text{O}} &= 1.4 \cdot 10^{-16} T \cdot 10^{2.53 - 17.2T^{-0.5}}, \\
 W_6^{\text{N}_2} &= 5.2 \cdot 10^{-15} T^{1.5} \exp(-37T^{-0.5}), \\
 W_6^{\text{He}} &= 5.6 \cdot 10^{-16} \exp(-50T^{-0.5}). \quad (7)
 \end{aligned}$$

d) Gasdynamic Jet Parameters

For a complete description of the kinetics it is necessary to solve simultaneously the system (2) and the equations of gasdynamics. The gasdynamic parameters of an expanding gas were determined in analogy with^[23], i.e., it was assumed that it is legitimate to use adiabatic motion of the medium with a certain effective adiabatic exponent γ . The density N and the temperature T of the gas are then given by

$$N = N_5 \left(1 + \frac{\gamma - 1}{2} M^2\right)^{-1/(\gamma - 1)}, \quad T = T_5 \left(1 + \frac{\gamma - 1}{2} M^2\right)^{-1}, \quad (8)$$

where N_5 and T_5 are the stagnation parameters, $M = V/a$ is the local value of the Mach number, $a = \sqrt{\gamma RT/\mu}$ is the local velocity of sound, R is the gas constant, and $\mu = \sum \mu_i \gamma_i$ ($i = \text{N}_2\text{O}, \text{N}_2, \text{He}$) is the molecular weight of the mixture. As in^[23], we used a Mach-number field calculated by the method of characteristics for the escape of a gas from a slit into vacuum. Some results of this calculation are given in^[23]. For γ we chose the expression

$$\gamma = \left(\frac{5}{2} + \sum_M \gamma_M\right) / \left(\frac{5}{2} + \sum_M \gamma_M\right), \quad (9)$$

where the summation is over the molecular components.

e) Inversion and Gain

The purpose of the solution of the system (2) was to obtain the distributions of the population difference ΔN of the level $00^01 - 10^00$ of the N_2O molecule as the distance from the slit is varied. The inversion was then determined from the relation

$$\Delta N = N \gamma_{N,0} (y_3 - y_2^2) (1 - y_2)^2 (1 - y_2^2) (1 - y_3), \quad (10)$$

where

$$y_i = \exp\left(-\frac{h\nu_i}{kT_i}\right), \quad T_i = -\frac{h\nu_i}{k} \left[\ln \frac{\epsilon_i}{r_i + \epsilon_i}\right]^{-1}.$$

One of the most important directly-measured characteristics of lasers, in addition to ΔN is the gain α for the vibrational-rotational transition. It was calculated by the known formula

$$\alpha = \frac{c^2 A}{8\pi\nu^2} g_n \left(\frac{N_m}{g_m} - \frac{N_n}{g_n}\right) F(\nu - \nu_0), \quad (11)$$

where ν is the transition frequency, $A = 0.278 \text{ sec}^{-1}$ ^[24] is the Einstein coefficient, c is the speed of light, $N_m(n)$ is the population of the vibrational rotational level, $g_m(n)$ is its statistical weight, $F(\nu - \nu_0)$ is the line-shape factor, and ν_0 is the frequency at the center of the line. If the line width is determined both by the Doppler effect and by collisions with other gas molecules, the expression for F at the line center ($\nu = \nu_0$) takes the form^[10]:

$$F = \frac{2}{\Delta\nu_D} \sqrt{\frac{\ln 2}{\pi}} H(b). \quad (12)$$

The vibrational-rotational inversion $\Delta N_{V,J}$ was determined from the formula

$$\Delta N_{V,J} \approx \frac{hcB}{kT} (2J+1) \exp\left[-\frac{hcB}{kT} J(J+1)\right] \Delta N, \quad (13)$$

where J is the rotational quantum number and B is the rotational constant (its value $\approx 0.42 \text{ cm}^{-1}$ is the same, with good accuracy^[11] for both the upper level 00^01 and the lower level 10^00). The cross sections of the broadening collisions were taken to be^[24]

$$\begin{aligned}
 \sigma_{\text{N}_2\text{O}-\text{N}_2\text{O}} &= 7 \cdot 10^{-13} \text{ cm}^2, \quad \sigma_{\text{N}_2\text{O}-\text{N}_2} = 7.7 \cdot 10^{-13} \text{ cm}^2 \\
 \sigma_{\text{N}_2\text{O}-\text{He}} &= 3.6 \cdot 10^{-13} \text{ cm}^2
 \end{aligned}$$

3. SOLUTION AND PRINCIPAL CALCULATION RESULTS

The system (2) with allowance for relations (3)–(9) was solved with a computer by the Runge-Kutta method. The initial conditions presuppose complete thermodynamic equilibrium in the gas ($T_2 = T_3 = T_4 = T$) with temperature $T_{\text{init}} = 2T_5/(\gamma + 1)$ and density $N_{\text{init}} = N_5(2/(\gamma + 1))^{1/(\gamma - 1)}$, (i.e., $M = 1$ in the critical section of the slot). The solution was obtained for different temperatures T_5 , pressures P_5 , slot widths h , and for different chemical compositions. We calculated the changes in the total vibrational inversion, in the gain, and in the vibrational and gas temperatures as functions of the distance from the slot. The gain was calculated for the P(20) line of the (00^01)–(10^01) transition corresponding to the transition in the sounding laser.

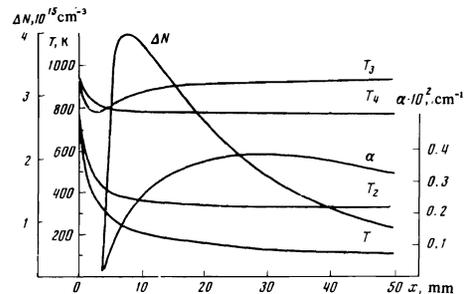


FIG. 2. Distributions of the inversion ΔN , of the gain for the line P(20), and of the vibrational and gas temperatures along the outflow axis. Mixture $\text{N}_2\text{O}:\text{N}_2$ (1:4); $T_5 = 1200^\circ\text{K}$, $P_5 = 25 \text{ atm}$. Width of slot $h = 0.08 \text{ cm}$.

Figure 2 shows typical results of the calculation of the distributions of the inversion ΔN , of the gain α , and of the vibrational and gas temperatures. The vibrational temperatures T_4 and T_2 and the gas temperature T first fall off abruptly, after which T_2 and T_4 "freeze" while T continues to decrease. The temperature T_3 reaches a minimum at $x \approx 2$ mm and then begins to increase. The growth of T_3 , due to the rapid nonresonant exchange N_2 of the level (00^01) with $(v=1)^{[25]}$ improves the conditions for inversion. Inversion is produced when a definite gap is reached between T_3 and T_2 (T_3/T_2 must be ≈ 1.89). The inversion reaches a maximum at $x \approx 7$ mm. The gain α first increases rapidly up to $x \approx 20$ mm and subsequently varies slowly. The presence of N_2 causes an increase of T_3 . With decreasing N_2 content in the mixture, the growth of T_3 slows down (there is no increase of T_3 for the $N_2O:He$ mixture).

The results of the calculations have demonstrated the usefulness of N_2O as a working molecule for GDL and have served as a justification of the experiment.

4. DESCRIPTION OF EXPERIMENT

The inverted population in N_2O GDL was investigated by the weak-signal amplification method using the setup described in detail earlier^[26] and employed in a number of studies of CO_2 GDL. It comprised a quasistationary shock-tube GDL. The mixture was heated by a reflected shock wave. The escape was through a short slot nozzle (width $h = 0.08$ cm, aperture angle 120°).

For the probing we used an N_2O gas-discharge laser^[27] operating on the P(20) line of the $(00^01) \rightarrow (10^00)$ transition (a diffraction grating was used in the resonator). The laser power was ~ 0.5 W. The zero-signal level was registered with the aid of a shutter prior to the experiment and was verified after the experiment. The temperature and pressure ($T_0 = T_5$, $P_0 = P_5$) of the working mixture behind the reflected wave prior to the escape was calculated from the shock-wave velocity measured with the aid of contact pickups. The velocity measurement accuracy was $\pm 5\%$, and that of the pressure and temperature $\pm 7-10\%$.

Figure 3 shows typical oscillograms of the intensity of the sounding beam passing through the working mixture 20 mm downstream from the critical section. The Schlieren peak (arrow F), which is clearly seen on all the oscillograms, is due to the passage, through the optical axis, of the shock front propagating in the receiver

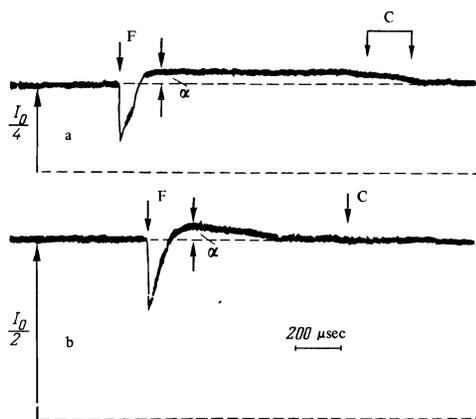


FIG. 3. Oscillograms of the experiments for the mixture $N_2O:N_2$ (1:4). a) $T_5 = 1200^\circ K$, $P_5 = 24$ atm, $\alpha = 3.8 \times 10^{-2}$ over 9 cm; b) $T_5 = 1630^\circ K$, $P_5 = 20$ atm, $\alpha = 3.0 \times 10^{-2}$ over 9 cm.

after the start of the escape of the mixture through the slot. The peak is followed by an increase of the signal, which passes through the zero level I_0 and then is amplified. The amplification continues for about 600–1000 μsec (the arrows C), decreasing smoothly to zero. The duration of the amplification depends mainly on the characteristics of the employed shock-tube construction (the lengths of the compartments etc.). The accuracy with which the gain α was measured was ± 20 to 30%.

5. EXPERIMENTAL RESULTS AND COMPARISON WITH CALCULATION

a) Measurement of the Gain Along the Jet Axis

To perform the experiments it was first necessary to find the distance from the critical section of the slit to the probing axis at which the gain is maximal or nearly so. To this end a number of experiments were performed to determine the dependence of α on x for different mixtures, starting from the data obtained by the calculation described above. Figure 4 shows the theoretical and experimental results of the investigation of $\alpha(x)$ for the mixtures $N_2O:N_2(1:8)$ and $N_2O:N_2:He(1:2:7)$. The calculation agrees satisfactorily with the experimental values of α . It is seen from Fig. 4 that at a distance $x \approx 20$ mm the values attained by α are close to the maximal α_{max} for both mixtures. In the same setup, in the case of the CO_2 GDL, the distances at which α reaches a maximum were approximately the same and varied little under various conditions. For this reason the probing in all the succeeding experiments was carried out at a distance $x = 20$ mm from the critical section.

b) Results of Investigation of the $N_2O:N_2$ Mixture

The simplest gas mixture investigated in detail in the present study was $N_2O:N_2$. The presence of inversion for the binary $N_2O:N_2$ mixture was established by us for the first time.

Figure 5 shows the dependence of the gain on the ratio of the components of the mixture $N_2O:N_2$. All the experimental values shown in Fig. 5 were obtained at a fixed partial N_2O pressure in the mixture, $P_5^{N_2O} = 4.4$ atm, and at a temperature $T_5 = 1200 \pm 70^\circ K$. In this series of experiments, when the values of $\kappa_{N_2} = [N_2]/[N_2O]$ range from zero to 8, the stagnation pressure P_5 ranged accordingly from 4.4 to 39.6 atm. It was established that an inversion occurs in the case of escape of pure N_2O ($\alpha_{absorb} = -2.8 \times 10^{-2} cm^{-1}$). It is also seen that when κ_{N_2} increases from 2 to 4, the gain increases rapidly to $\alpha \approx 0.4 \times 10^{-2} cm^{-1}$. Further, at $\kappa_{N_2} > 4$, its growth slows down. This character of the dependence of the gain on the composition of the mixture can be attributed to the presence of two competing processes, namely, the increase of the energy stored at the upper laser level as a result of exchange with the vi-

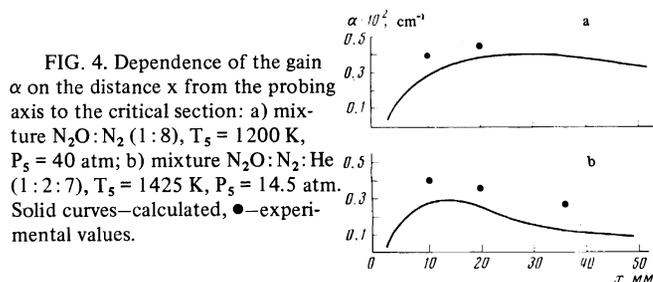


FIG. 4. Dependence of the gain α on the distance x from the probing axis to the critical section: a) mixture $N_2O:N_2(1:8)$, $T_5 = 1200$ K, $P_5 = 40$ atm; b) mixture $N_2O:N_2:He(1:2:7)$, $T_5 = 1425$ K, $P_5 = 14.5$ atm. Solid curves—calculated, \bullet —experimental values.

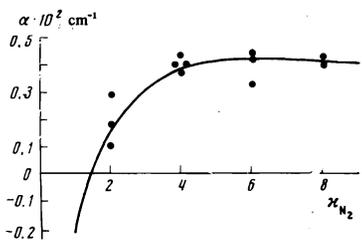


FIG. 5. Dependence of the gain α on the nitrogen content $\kappa N_2 = [N_2]/[N_2O]$ for the $N_2O:N_2$ mixture at constant values of the partial pressure $P_5 N_2O = 4.4$ atm and for constant temperature $T_5 = 1200 \pm 70^\circ K$. Solid curve—calculation, ●—experiment.

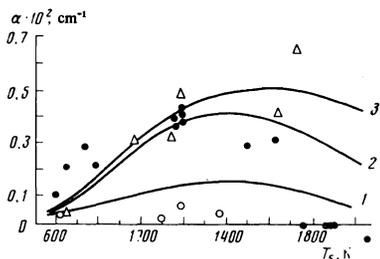


FIG. 6. Dependence of the gain α on the temperature T_5 for various pressures in the case of $N_2O:N_2(1:4)$: 1—theoretical curve for $P_5 = 12$ atm, ○—experimental values for $P_5 = 12 \pm 2$ atm; 2—theoretical curve for $P_5 = 25$ atm, ●—experimental values for $P_5 = 25 \pm 5$ atm; 3—theoretical curve for $P_5 = 45$ atm, △—experimental values at $P_5 = 45 \pm 5$ atm.

brationally-excited N_2 , and on the other hand the increase of the relaxation rate of the energy of this level as a result of the increase of the total pressure. Figure 5 shows also a theoretical plot of α against κN_2 (solid line), which agrees well with the experimental results. As seen from Fig. 5, the value $\kappa N_2 = 4$ is close to the optimal value. We have therefore carried out the subsequent detailed investigation of the $N_2O:N_2$ mixture for a component ratio 1:4.

Figure 6 shows the calculated and experimental plots of the gain α against the stagnation temperature for three values of the pressure P_5 (12, 25, and 45 atm). The maxima of the calculated curves shift towards higher temperatures with increasing pressure P_5 . The dependence of α on the pressure is discussed below. We note here that when the pressure increases from 25 to 45 atm the value of α changes little, although it does increase. As seen from Fig. 6, for the temperature interval 600–1000°K the optimal pressure is $P_5 \approx 25$ atm. At a given pressure, the experimental values of α exceed the calculated ones (by approximately 2–3 times). It must be emphasized that in the operating-temperature region $T < 300^\circ K$ there are no experimental data whatever for the relaxation rates in N_2O . This temperature region, however, is most important for GDL, inasmuch as in this region the gas temperature T decreases in the case of fast adiabatic expansion to a temperature much lower than 300°K. As shown by investigations (cf.^[15]), at $T \leq 300^\circ K$ the decrease of the relaxation rate with decreasing temperature is much slower than in the region $T > 300^\circ K$. Therefore, to attain satisfactory agreement between the calculated and experimental data at low temperatures it was assumed that W_8^M at $T \leq 300^\circ K$ remains unchanged.

From the practical point of view it is important that inversion and noticeable amplification are registered already at $T_5 \approx 600^\circ K$. (In the case of a CO_2 GDL, judging from the literature, inversion sets in only at $T_5 > 800^\circ K$ and in the presence of He or H_2O in the mixture.)

For $T_5 > 1700^\circ K$, the available experimental points indicate a jumplike vanishing of the gain, thus contradicting the calculations. This fact is easy to explain—in our calculation we did not take into account the N_2O dissociation, which at temperatures on the order of 1700°K greatly decreases the number of working molecules. In addition, it appears that the dissociation products (NO , O , etc.) are very effective in the vibrational de-activation of the N_2O molecules. The strong dependence of the dissociation rate on the temperature serves as an indirect indication of the chain character of the process of N_2O dissociation (energy chain).

Figure 7 shows plots of the gain against the pressure P_5 for three different temperatures (750, 1200, and 1600°K) in the case of the mixture $N_2O:N_2(1:4)$. The results of the calculation are in agreement with experiment, so that it can be concluded that in the indicated pressure intervals we have attained the optimal values P_5^{opt} in the experiment.

Calculation^[10] has shown that the optimal temperature for the inversion at various pressures is $T_5 \approx 1100$ – $1200^\circ K$. We note that in the case of CO_2 GDL, in the presence of He or H_2O in the mixture, close values of the inversion ($\Delta N \approx 10^{16} \text{ cm}^{-3}$, $T_5 = 1200^\circ K$, $P_5 = 60$ atm) are attained at much higher temperatures, $T_5 > 1800^\circ K$.

c) Results of the Investigation for Helium-Containing Mixtures $N_2O:He(1:3)$, $N_2O:N_2:He(1:2:7)$, $N_2O:CO:He(1:2:7)$

The question of whether an N_2O GDL can operate with a binary $N_2O:He$ mixture, and if this is the case, the question of the attainable gain, are both of interest for the completion of the general picture of the relaxation mechanism. This mixture was not investigated by us in detail, but a number of experiments were performed, in which gain was observed for $N_2O:He(1:3)$. Thus, for example, at $P_5 \approx 10$ atm and $T_5 \approx 1600^\circ K$, the gain was $\alpha \approx 0.25 \times 10^{-2} \text{ cm}^{-1}$. We note that for this mixture, too, amplification was obtained for the first time.

We investigated the dependence of the gain α on T_5 and P_5 for the mixture $N_2O:N_2:He(1:2:7)$. Figure 8 shows plots of the gain α against the temperature T_5 for pressures P_5 equal to 14.5 and 45 atm. When the pressure is increased in the experiment from 14.5 to 45 atm the gain increases abruptly, reaching at $T_5 \approx 1550^\circ K$ and $P_5 \approx 45$ atm a relatively large value $\alpha \approx 1 \times 10^{-2} \text{ cm}^{-1}$. At $P_5 = 14.5$ atm, the results of the calculations were in satisfactory agreement with the experimental data, and at $P_5 = 45$ atm the calculated values were half as large as the experimental ones and, most importantly, the calculation did not result in so abrupt an increase of the

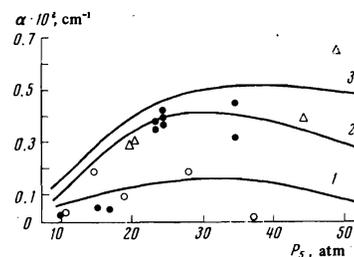


FIG. 7. Dependence of the gain α on the pressure P_5 for various temperatures in the case $N_2O:N_2(1:4)$: 1—theoretical curve for $T_5 = 750^\circ K$, ○—experimental values at $T_5 = 750 \pm 100^\circ K$; 2—theoretical curve for $T_5 = 1200^\circ K$, ●—experimental values at $T_5 = 100 \pm 100^\circ K$; 3—theoretical curve for $T_5 = 1600^\circ K$, △—experimental values at $T_5 = 1600 \pm 100^\circ K$.

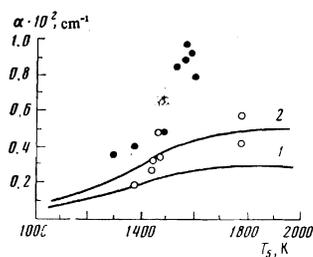


FIG. 8

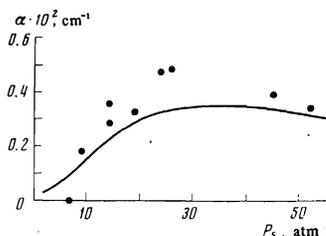


FIG. 9

FIG. 8. Dependence of the gain α on the temperature for various pressures. Mixture $\text{N}_2\text{O}:\text{N}_2:\text{He}$ (1:2:7). 1—theoretical curve for $P_5 = 14.5$ atm, \circ —experimental values at $P_5 = 14.5 \pm 2$ atm; 2—theoretical curve for $P_5 = 45$ atm, \bullet —experimental values at $P_5 = 45 \pm 6$ atm.

FIG. 9. Gain α vs. pressure for the mixture $\text{N}_2\text{O}:\text{N}_2:\text{He}$ (1:2:7). Theoretical curve for $T_5 = 1420$ K, \bullet —experimental values at $T_5 = 1420 \pm 50^\circ\text{K}$.

gain as the temperature increased from 1300 to 1600°K. This may mean that temperature dependences chosen for the relaxation rate in N_2O in the presence of He have not been quite correctly chosen.

Figure 9 shows the theoretically and experimentally obtained plots of the gain α against the pressure P_5 at a temperature $T_5 = 1420^\circ\text{K}$. As the pressure is increased from 10 to 20 atm, as seen from Fig. 9, the gain increases rapidly, reaching a maximum at $P_5 \approx 30$ atm, and subsequently decreases slowly. The calculation describes the experimental results approximately correctly.

We also calculated the inversion as a function of the pressure P_5 at the optimal temperature $T_5 = 1200^\circ\text{K}$ ^[10] For the mixture $\text{N}_2\text{O}:\text{N}_2:\text{He}$ (1:2:7), calculation yields a value $\Delta N_{\text{max}} \approx 2.3 \times 10^{16} \text{ cm}^{-3}$ at the optimal pressure $P_5 \approx 100$ atm. This is larger than in CO_2 GDL with an analogous mixture composition.

To analyze the role of the nonresonant exchange in an N_2O laser, it is of interest to trace the influence of replacement of N_2 by CO on the magnitude of the inversion, since the energy defect between the levels (00^0_1) of N_2O and ($V = 1$) of CO is comparable in magnitude ($\Delta E_{\text{CO}} = 80 \text{ cm}^{-1}$), but the ($v = 1$) CO level is lower than 00^0_1 . In the calculation for the $\text{N}_2\text{O}:\text{CO}:\text{He}$ (1:2:7) mixture it was assumed that the rate of the V - T relaxation is determined mainly by the helium, since its content in the mixture is large, and that CO exerts the same influence on the relaxation of the upper level 00^0_1 as N_2 . The probability of exchange of the levels (00^0_1) and $\text{CO}(v = 1)$ was taken from^[14]:

$$W^{(v)} = 10^{-16} T \left[\frac{150}{1+y} - \frac{20}{(2/y+1)} + 50 \right] \exp\left(-\frac{115}{T}\right),$$

Calculation^[10] yields lower maximum values of ΔN than for nitrogen-containing mixtures. This can be attributed precisely to the fact that in the presence of N_2 an important role is assumed by the growth of T_3 due to the fast nonresonant exchange. No experiments were performed aimed at observing amplification in the $\text{N}_2\text{O}:\text{CO}:\text{He}$ mixture.

Interest attaches to the investigation of the influence of water vapor on the operation of an N_2O GDL. In a CO_2 GDL, even small additions of water greatly influence the inversion, and a water-vapor concentration $[\text{H}_2\text{O}] = 1$ to 2% is optimal. In the N_2O GDL, appreciable inversions and gains are obtained with $\text{N}_2\text{O}:\text{N}_2$ mixtures even without water. It was established experimentally that addition of water vapor does not increase the gain appreciably^[10].

6. CONCLUSION

As is already noted, so far there are only several studies in which gain and lasing was established in GDL operating on mixtures containing N_2O . We have presented above the first theoretical analysis of the operation of an N_2O GDL for a concrete experimental setup. Amplification was obtained in experiment in the mixtures $\text{N}_2\text{O}:\text{N}_2$, $\text{N}_2\text{O}:\text{He}$, $\text{N}_2\text{O}:\text{N}_2:\text{He}$, and $\text{N}_2\text{O}:\text{N}_2:\text{H}_2\text{O}$. The results of the developed calculation procedure are in accord with experiment in a wide range of parameters. We investigated in detail the laser operating mechanism. A new physical effect in comparison with the CO_2 GDL is the appreciable growth of the temperature T_3 (which determines the population of the upper laser level) during the course of the expansion of the mixture, owing to the fast nonresonant exchange of the levels (00^0_1) N_2O and ($v = 1$) N_2 . It appears that at present the role of nonresonant exchange in the analysis of the possibilities of producing effective lasers is still underestimated. Thus, investigations of the $\text{N}_2\text{O}:\text{N}_2$ mixture have shown that if it is used, in contrast to the $\text{CO}_2:\text{N}_2$ mixture, an appreciable inversion and an appreciable gain are obtained. The optimal component ratio for the $\text{N}_2\text{O}:\text{N}_2$ mixture is 1:4. The addition of a small amount of water to the mixture $\text{N}_2\text{O}:\text{N}_2$ (1:4) increases somewhat the gain. The fact that effective operation can be obtained with the $\text{N}_2\text{O}:\text{N}_2$ mixture, which was established here for the first time, is of great importance for the practice, since the mixtures $\text{N}_2\text{O}:\text{N}_2$ and $\text{N}_2\text{O}:\text{N}_2:\text{H}_2\text{O}$ are relatively inexpensive.

Experiments yielded a high gain $\alpha \approx 1 \times 10^{-2} \text{ cm}^{-1}$ for the mixture, $\text{N}_2\text{O}:\text{N}_2:\text{He}$ (1:2:7). The inversion $\Delta N_{\text{max}} \approx 10^{16} \text{ cm}^{-3}$ is somewhat higher than in the CO_2 GDL and is attained at a much lower temperature $T_5 \approx 1200^\circ\text{K}$, whereas for CO_2 the optimum is at $T_5 > 1800^\circ\text{K}$. This is of practical importance, since the optimal temperatures for N_2O GDL lie in a region that is well managed by modern technology. In addition, the lowering of T_5 will apparently make it possible to increase the efficiency of N_2O GDL as compared with CO_2 GDL.

An advantage of the N_2O laser is also the fact that its emission ($\lambda \approx 10.9 \mu$), which lies in the transparency window of the atmosphere, is less absorbed than that of the CO_2 laser^[28], since the content of N_2O in the atmosphere is much lower than that of CO_2 .

On the whole, the results of the investigation indicate that N_2O is a useful working medium for GDL.

¹G. Herzberg, Spectra of Diatomic Molecules, Van Nostrand [Russ. transl. IIL, 1949].

²C. K. N. Patel, Appl. Phys. Lett., 6, 12 (1965).

³C. K. N. Patel, Appl. Phys. Lett., 8, 69 (1966).

⁴A. Sugiyama, and H. Yuaba, Phys. Lett., 28A, 120 (1968).

⁵B. R. Bronfin, L. R. Boedeker and J. P. Cheyer, Appl. Phys. Lett., 16, 214 (1970).

⁶Yu. A. Grin', V. M. Polyakov, and V. G. Testov, ZhETF Pis. Red. 18, 260 (1973) [JETP Lett. 18, 155 (1973)].

⁷V. F. Gavrikov, A. P. Dronov, V. K. Orlov, and A. K. Piskunov, Kvantovaya elektronika, No. 5, 119 (1973) [Sov. J. Quant. Electr., 3, (1974)].

⁸A. S. Biryukov and B. F. Gordiets, Prikl. Mat. Teor. Fiz. 6, 29 (1972); FIAN Preprint No. 32 (1972).

⁹A. S. Biryukov and L. A. Shelepin, Zh. Tekh. Fiz. 44, 1232 (1974) [Sov. Phys.-Tech. Phys. 19, 775 (1974)]; FIAN Preprint No. 59 (1973).

¹⁰A. S. Biryukov, A. Yu. Volkov, A. I. Demin, E. M. Kudryatsev, Yu. A. Kulagin, N. N. Sobolev, and L. A.

- Shelepin, FIAN Preprint No. 140 (1974).
- ¹¹A. S. Biryukov, V. K. Konyukhov, A. I. Lukovnikov, and R. I. Serikov, *Zh. Eksp. Teor. Fiz.* **66**, 1248 (1974) [*Sov. Phys.-JETP* **39**, 610 (1974)]; FIAN Preprint No. 9 (1973).
- ¹²R. N. Schwartz, Z. J. Slawsky, and K. F. Herzfeld, *J. Chem. Phys.*, **20**, 1591 (1952); R. N. Schwartz and K. F. Herzfeld, *J. Chem. Phys.*, **22**, 767 (1954).
- ¹³C. B. Moore, R. E. Wood, Bei-Lok Hu, and J. T. Yardley, *J. Chem. Phys.* **46**, 4222 (1967); A. S. Biryukov, R. I. Serikov, and E. S. Trekhov, *Zh. Eksp. Teor. Fiz.* **59**, 1513 (1970) [*Sov. Phys.-JETP* **32**, 825 (1971)].
- ¹⁴M. Margottin-Maclou, H. Gueguen, L. Doyennette, and L. Henry, *C. R., Paris*, **274**, 1414-B, (1972).
- ¹⁵H. Gueguen, A. Carion, M. Margottin-Maclou, L. Doyennette, *C. R., Paris*, **274**, 482-B (1972).
- ¹⁶C. J. S. M. Simpson, K. B. Bridgman and T. R. D. Chandler, *J. Chem. Phys.*, **49**, 509 (1968).
- ¹⁷J. F. Roach and W. R. Smith, *J. Chem. Phys.*, **50**, 4114 (1969).
- ¹⁸G. J. Mullaney, H. G. Ahlstrom and W. H. Christiansen, *IEEE, J. of Quant. Elect.*, **QE-7**, 551 (1971).
- ¹⁹J. T. Yardley, *J. Chem. Phys.*, **49**, 2816 (1968).
- ²⁰W. Griffith, D. Brickl and V. Blackman, *Phys. Rev.*, **102**, 1209 (1956).
- ²¹M. Huetz, P. Chevalier and B. Sanson, *Astronautica Acta*, **17**, 645 (1972).
- ²²R. L. Taylor, S. Bitterman, *Rev. Mod. Phys.*, **41**, 26 (1969).
- ²³E. M. Kudryavtsev and V. N. Faizulaev, FIAN Preprint No. 68 (1972).
- ²⁴N. Djeu, T. Kan and G. Wolga, *IEEE, J. of Quant. Elect.*, **QE-4**, 783 (1968).
- ²⁵B. F. Gordiets, A. I. Osipov, E. V. Stupochenko, and L. A. Shelepin, *Usp. Fiz. Nauk* **108**, 655 (1972) [*Sov. Phys.-Uspekhi* **15**, 759 (1973)].
- ²⁶A. P. Dronov, A. S. D'yakov, E. M. Kudryavtsev, and N. N. Sobolev, *ZhETF Pis. Red.* **11**, 516 (1970) [*JETP Lett.* **11**, 353 (1970)].
- ²⁷A. I. Demin, E. M. Kudryavtsev, Yu. A. Kulagin, and N. N. Sobolev, *Kvantovaya elektronika* **1**, 2499 (1974) [*Sov. J. Quant. Electr.* **1**, 1393 (1975)].
- ²⁸U. P. Oppenheim and P. Melman, *JOSA*, **60**, 332 (1970).

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