

*EFFECT OF TEMPERATURE ON THE PROPERTIES OF PULSED LASER ACTION ON ELECTRON TRANSITIONS IN DIATOMIC MOLECULES*

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The temperature dependence of the optical gain is studied theoretically. A detailed calculation of the optical gain for the  $C^3\Pi_u \rightarrow B^3\Pi_g$  transition in the nitrogen molecule, corresponding to ultraviolet laser action, is made at temperatures between 400 and 50°K for two limiting cases of (fast and slow) rotational relaxation. It is shown that a redistribution of the laser power among the rotational lines and a significant increase in the optical gain should take place upon cooling. It is established experimentally that, in accord with the calculations, the maximum of the laser power output shifts toward lower J on cooling, while the optical gain increases significantly. At low temperatures, a new oscillation on the 1 → 0 band (edge at 3159 Å) of the same band system in nitrogen is detected. Its rotational structure is identified. When the gas is cooled to a temperature near that of liquid nitrogen, the total laser output of the ultraviolet oscillation increases by a factor of ~ 1.5, and for the infrared by a factor of 1.35. The laser power output in the weak bands increases to a much greater extent. The excitation mechanisms for ultraviolet and infrared oscillation in molecular nitrogen are discussed on the basis of the temperature dependences obtained.

1. INTRODUCTION

It has been shown recently that the temperature of the working gas significantly affects the operation of gas lasers. In particular, it has been shown that the power output of a helium-neon laser decreases upon cooling.<sup>[1-4]</sup> On the other hand, it has been shown that a decrease in the temperature of the working gas improves the operation of lasers in the vibrational-rotational transitions in molecules.<sup>[5,6]</sup> A theoretical analysis of the effect of temperature on the optical gas in vibrational-rotational transitions was presented by Patel.<sup>[7]</sup> The effect of temperature on the operation of other types of gas lasers remains virtually unstudied.

There is great interest in the study of the effect of the operating gas temperature on the properties of the laser output, for lasers using the electron transitions in diatomic molecules. As is well known, the highest peak power output is achieved by such lasers, reaching 1 MW.<sup>[8,9]</sup> On the other hand, on the basis of data on the temperature dependence on the vibrational-rotational transitions, one can also expect a strong temperature dependence for generation by electronic transitions in molecules.

In the present research, we studied the effect of temperature on the properties of laser action in electronic transitions in nitrogen molecules.<sup>[8-16]</sup> The principal attention is paid to ultraviolet oscillation. Some preliminary results were published by us earlier.<sup>[17]</sup>

2. THEORETICAL DISCUSSION OF THE OPTICAL GAIN

Further discussion is carried out under the following assumptions: 1) the molecule is regarded as a rigid symmetric top;<sup>[18]</sup> 2) the case of Hund coupling is achieved; 3) the wave function of the molecule can be

represented in the form of the product of electronic, vibrational, and rotational wave functions.

Under these assumptions, the gain at the center of the Doppler-broadened line corresponding to an electronic-vibrational-rotational transition from the upper level with quantum numbers  $v'J'$  to the lower level  $v''J''$  can be written in the form

$$k(v_0) = Q_{v''v'} \frac{B_{v''} N_{v''} S_{J'J''}}{T_{rot}'' \sqrt{T_{mol}}} \left\{ a \frac{T_{rot}'' \exp[-B_{v''}hcJ'(J'+1)/kT_{rot}']}{T_{rot}' \exp[-B_{v''}hc\Omega'(\Omega'+1)/kT_{rot}']} - \frac{\exp[-B_{v''}hcJ''(J''+1)/kT_{rot}'']}{\exp[-B_{v''}hc\Omega''(\Omega''+1)/kT_{rot}']'} \right\},$$

$$Q_{v''v'} = \frac{4\pi^3 c S_e(\bar{r}_{v''v'}) q_{v''v'}}{3k \sqrt{2\pi k/M}}, \quad a = \frac{B_{v''} N_{v''}}{B_{v'} N_{v'}}, \quad (1)$$

where the quantities denoted by a single prime refer to the upper operating state, and those with two primes, to the lower;  $S_e(\bar{r}_{v''v'})$  is the electronic transition strength as a function of the r-centroid,  $q_{v''v'}$  is the Franck-Condon factor for the band  $v' \rightarrow v''$ ,  $S_{J'J''}$  is the rotational line strength or Henle-London factor,<sup>[18,19]</sup>  $N_v$  is the total population of the vibrational level,  $B_v$  the rotational constant,  $\Omega$  the total electronic angular momentum of the molecule,  $h$  Planck's constant,  $k$  Boltzmann's constant,  $c$  the velocity of light, and  $M$  the molecular weight. In what follows, we shall call the quantity  $a$  the coefficient of vibrational inversion. In the derivation of the formula it was assumed that the population distribution over the rotational sublevels of each vibrational level is a Boltzmann one with a temperature  $T_{rot}$ , while  $T_{mol}$  describes the distribution of molecules over the velocities, and enters into the expression for the Doppler line width.

The formula given does not differ strongly from the formula obtained in<sup>[7]</sup> for vibrational transitions. This is connected with the fact that even for electronic tran-

sitions, the decisive factors are the redistribution of the population over the rotational sublevels upon change in temperature, and the change in the Doppler line width. The difference between the formulas is primarily that the denominators  $\exp[-B_V hc \Omega(\Omega + 1)/kT_{\text{rot}}]$  appear and describe the effect of the angular momentum. Furthermore, electronic transitions possess certain other characteristic features. In particular, for electronic transitions, the rotational constants  $B_{V'}$  and  $B_{V''}$  of the upper and lower operating states can be significantly different from one another. However, the most serious differences can be associated with the value of the coefficient of vibrational inversion  $a$ , which is fundamentally determined by the ratio  $N_{V'}/N_{V''}$ .

The populations of the vibrational levels  $N_{V'}$  and  $N_{V''}$  are in turn determined by the conditions of excitation and decay of the levels. For example, if we assume that the excitation of all the levels considered takes place by direct electron impact from the ground state, and that the population of the ground state is constant, while the decay of the levels is determined by spontaneous emission, then it can be concluded that  $N_{V'}$  and  $N_{V''}$  will not depend on the temperature of the gas. On the other hand, in the case when collisions of molecules with molecules play an important part in the process of excitation or decay of the operating levels, the populations  $N_{V'}$ ,  $N_{V''}$  are shown to be temperature dependent in the general case, since the collision cross sections and the rate of molecular collisions depend on the temperature. Thus, in addition to the explicit temperature dependence shown in (1), the gain can also depend on the temperature through the ratio  $N_{V'}/N_{V''}$ . The character of the dependence is determined by the mechanism of population and depopulation and cannot be described for the general case. The vibrational and electronic transitions can differ widely in this respect.

Specific calculations are given in the present paper for the P-branches of the transition  $C^3\Pi_u \rightarrow B^3\Pi_g$  of a nitrogen molecule (the second positive system of the band), corresponding to ultraviolet laser action. They are made under the assumption that  $N_{V'}/N_{V''}$  does not depend on the gas temperature, since, for this laser action, all the data indicate<sup>[12,13,20,21]</sup> that the principal role in the excitation of the operating levels is played by direct electronic impacts. Only the P-branches are considered, since one can show that, for a given electronic transition, the gain coefficient is largest for them. A more detailed discussion of the mechanisms of creation of inversion, both for ultraviolet and for infrared laser action of a nitrogen molecule is given below in the discussion of the results.

As a consequence of the fact that the duration of ultraviolet laser action amounts to 5–20 nanoseconds,<sup>[8,9,11,12]</sup> the radiation lifetime of the upper working level  $C^3\Pi_u$  is about  $4 \times 10^{-8}$  nanosecond<sup>[22,23]</sup> while the rotational relaxation of the nitrogen molecule to its ground state<sup>[24,25]</sup> at room temperature and a pressure of 1 Torr amounts to about  $5 \times 10^{-7}$  sec, it can be expected that at low pressures and temperatures of the gas, during the time of the laser action, the rotational relaxation cannot take place (slow rotational relaxation), while at high temperatures and pressures, conditions can be encountered in which the time of rotational relaxation is comparable with or even shorter than the

time of existence of the inversion (fast rotational relaxation). In the latter limiting case, for all the vibrational levels considered a Boltzmann distribution is established over the rotational sublevels with a temperature  $T_{\text{rot}} = T_{\text{mol}}$ . In the first limiting case,  $T'_{\text{rot}}$  and  $T''_{\text{rot}}$  are determined by the mechanism of excitation of the levels and can differ from  $T_{\text{mol}}$ .

In direct excitation from the ground state of the molecule, the distribution over the rotational sublevels of the excited states, according to existing data<sup>[18]</sup>, duplicates the populational distribution for the ground state. This corresponds to  $T'_{\text{rot}} = T_{\text{mol}} B_V/B_0$  and  $T''_{\text{rot}} = T_{\text{mol}} B_{V''}/B_0$ , where  $B_0$  is the rotational constant for the ground state. Equation (1) for the gain takes in the case of slow rotational relaxation the form

$$k(v_0) = Q_{v'v''} \frac{B_0 N_{v''} S_{J',J''}}{T^{3/2}} \left\{ \frac{N_{v'} \exp\left[-\frac{B_0 hc}{kT} J'(J'+1)\right]}{N_{v''} \exp\left[-\frac{B_0 hc}{kT} \Omega'(\Omega'+1)\right]} \frac{\exp\left[-\frac{B_0 hc}{kT} J''(J''+1)\right]}{\exp\left[-\frac{B_0 hc}{kT} \Omega''(\Omega''+1)\right]} \right\},$$

where  $T = T_{\text{mol}}$ , and only the single rotational constant  $B_0$  enters into the exponent.

For the fast rotational relaxation, Eq. (1) remains practically unchanged, except that  $T'_{\text{rot}} = T''_{\text{rot}} = T_{\text{mol}} = T$ .

An analysis of the temperature dependence of the gain is conveniently started with the case in which  $N_{V'} \gg N_{V''}$ , i.e.,  $a \gg 1$ . In this case, the second terms in the curly brackets in Eqs. (1) and (2) can be discarded. Then the dependence of the gain on  $J'$  is the same as the dependence of the intensity of spontaneous emission from the upper level. For the  $P_1$ -th branch ( $\Omega' = \Omega'' = 0$ ) and for the case of fast relaxation we have

$$k(v_0) = Q_{v'v''} \frac{B_{v'} N_{v'}}{T^{3/2}} (J'+1) \exp\left[-\frac{B_{v'} hc}{kT} J'(J'+1)\right]. \quad (3)$$

It can be shown that the rotational quantum number  $J'_m$  corresponding to the maximum gain is determined approximately by the expression<sup>1)</sup>

$$J'_m \approx \sqrt{kT/2B_{v'} hc} - 3/2. \quad (4)$$

Then, for  $kT/B_{v'} hc \gg 1$  (this condition is usually well satisfied), it is not difficult to obtain the result

$$k(v_0, J'_m) \sim 1/T. \quad (5)$$

Thus, for large coefficients of vibrational inversion, the maximum gain is proportional to the reciprocal of the absolute temperature.

The cases of fast and slow rotational relaxations differ little when  $a \gg 1$ . Upon decrease of the vibrational inversion, it is not possible to discard the second term in (1) and (2), and the dependence of the gain on  $J'$  is changed. Figure 1 shows the calculated dependence of the gain on  $J'$  for the  $P_1$ -th branch ( $\Omega' = \Omega'' = 0$ ) for different temperatures. The solid and dashed curves represent slow and fast rotational relaxation. The curves are computed for  $a = 1$ . As is seen from the

<sup>1)</sup>For the case of a slow rotational relaxation, one must replace  $B_{v'}$  in (4) by  $B_0$ , which makes no difference, for the given transition, since  $B_{v'} = 1.826 \text{ cm}^{-1}$ , while  $B_0 = 2.010 \text{ cm}^{-1}$ .<sup>[18]</sup>

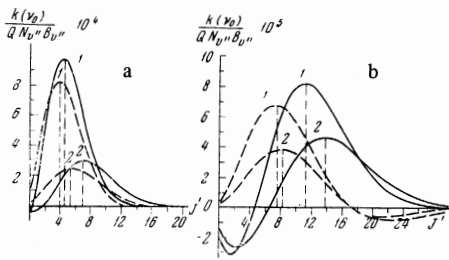


FIG. 1. Gain in relative units as a function of the rotational quantum number  $J'$  of the upper working level for gas temperatures: a - 1 - 75°K, 2 - 150°K, b - 1 - 300°K, 2 - 400°K. The continuous curves correspond to the case of slow rotational relaxation, the dashed to fast;  $a = N_V' B_V' / N_V'' B_V'' = 1$ .

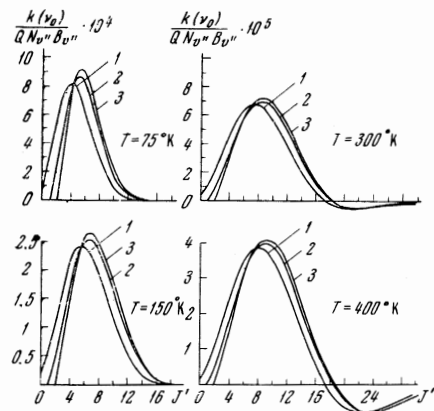


FIG. 4. The gain for different components of the multiplet as a function of the rotational quantum number  $J'$  of the upper working state for different gas temperatures. The curves denoted 1, 2, 3 refer to the branches  $P_1$  ( $\Omega = 0$ ),  $P_2$  ( $\Omega = 1$ ) and  $P_3$  ( $\Omega = 2$ ), respectively.

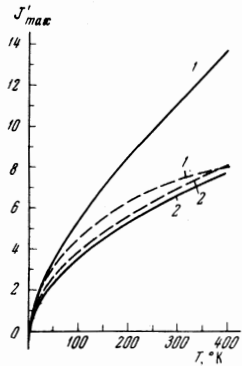


FIG. 2. Rotational quantum number  $J'_m$  of the upper working state, corresponding to a maximum value of the gain for  $a = 1$  (curve 1) and spontaneous emission (Curve 2). The continuous curves are for the slow rotational relaxation, dashed - the fast case.

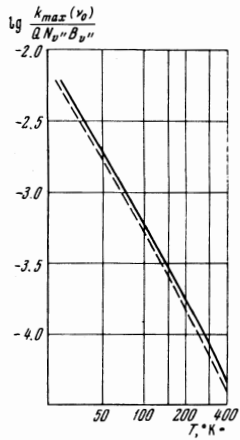


FIG. 3. Dependence of the maximum gain on the gas temperature. The continuous curve is for slow rotational relaxation, the dashed, for fast,  $a = 1$ .

drawing, with decrease in temperature, the maximum gain shifts in the direction of smaller  $J'$  and increases sharply in value. Whereas in the case  $a \gg 1$ , the dependence of the gain on  $J'$  differs very little for fast and slow rotational relaxations, these differences are already noticeable when  $a = 1$ . In particular,  $J'_m$  for these two cases differ substantially, especially at higher temperatures.

The dependence of  $J'_m$  on the temperature at  $a = 1$  is shown in Fig. 2 (curves 1). For comparison, curves 2 are shown for spontaneous emission from the upper level, i.e.,  $a \gg 1$ . The solid and dashed curves correspond to slow and fast rotational relaxation.

The dependence of the maximum gain on the temperature, also for  $a = 1$ , is shown in Fig. 3. The solid line corresponds to slow and the dashed to fast rotational relaxation. As can be seen from Fig. 3, the maximum

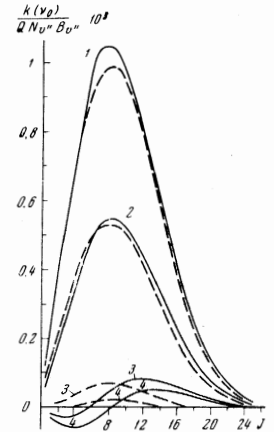


FIG. 5. The gain as a function of the upper rotational relaxation number for  $T = 300^\circ\text{K}$  for different values of the vibrational inversion  $a$ . Curve 1 -  $a = 2$ , 2 -  $a = 1.5$ , 3 -  $a = 1.0$ , 4 -  $a = 0.95$ .

gain in this case changes with temperature approximately as  $T^{-1.75}$ . Such an approximation in the temperature range from 400 to 50°K leads to an error in  $k(\nu_0)$  of not more than 5%. Thus, for  $a = 1$ , cooling of the gas from room temperature to the temperature of liquid nitrogen leads to an increase in the maximum gain of a factor of about 10.

The dependences given above were obtained for the  $P_1$  branch. As is well known from experiment,<sup>[14,16]</sup> laser action takes place principally on the branches  $P_1$ ,  $P_2$ , and  $P_3$ . The difference in the gain for branches  $P_1$ ,  $P_2$ , and  $P_3$  ( $\Omega' = \Omega''$  respectively equal to 0, 1 and 2) is shown in Fig. 4 for the case of fast rotational relaxation and  $a = 1$ .

Figure 5 gives the dependence of the gain on  $J'$  for different values of the quantity  $a$ . As before, the solid and dashed curves are for slow and fast rotational relaxation. It is seen from Fig. 5 that at small values of vibrational inversion ( $a \approx 1$ ) the differences between the cases of fast and slow rotational relaxations are considerable, while even for  $a = 1.5$ , these differences disappear almost completely. Upon further increase of  $a$ , the value of  $J'_m$  is virtually identical for both cases, while the value of the gain at the maximum is about 10% larger for the case of slow rotational relaxation than for fast rotational relaxation.

Thus, the theoretical calculation shows that cooling

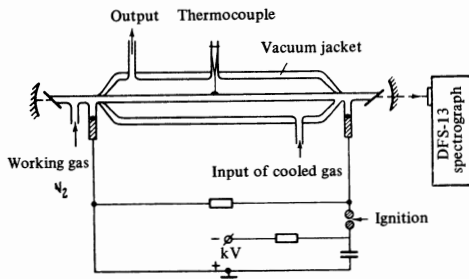


FIG. 6. Experimental apparatus, in which pumping of liquid nitrogen is employed for cooling

of nitrogen leads to a considerable increase of the gain for the transition  $C^3\Pi_u \rightarrow B^3\Pi_g$ , this increase being the more important the smaller the value of vibrational inversion. A similar behavior upon cooling should be expected also for other laser radiation by means of electronic transitions in diatomic molecules, if direct excitation by electrons from the ground state is the dominant process for the formation of inversion. The quantitative differences will be determined by the value of the vibrational inversion and the values of the rotational constants. The dependences of the gain on  $J'$  will also be somewhat different for the different branches.

### 3. EXPERIMENT

The experimental arrangement is pictured in Fig. 6. A laser of the ordinary type was used in the experiment, with external mirrors and with Brewster windows. The laser action was studied in two tubes with internal diameters 3 and 8 mm and active lengths of about 100 cm. The 3-mm tube was placed in a foam plastic tank with liquid nitrogen. The wider tube, shown schematically in Fig. 6, had a double jacket and was cooled by the passage of liquid nitrogen through it. The outer jacket was evacuated to produce the necessary thermal isolation. The temperature in this tube could be changed smoothly from room temperature to  $\sim 100^\circ\text{K}$ . The wall temperature of the working tube was measured by a thermocouple attached to its outer side. The measurements were made at low pulse repetition rates (2–4 Hz) and it was assumed that the gas temperature differed but little from the temperature of the wall. The tubes were excited by the discharge of a capacitor of  $0.01 \mu\text{f}$  by means of a controlled three-electrode discharge. The voltage on the capacitor reached 40 kV.

The output spectrum was studied on a DFS-13 diffraction spectrograph with a grating of 1200 lines per mm. The dispersion of the apparatus in first order amounted to  $2 \text{ \AA}/\text{mm}$ . The ultraviolet output was photographed principally in third order with a dispersion of  $\sim 0.5 \text{ \AA}/\text{mm}$ ; the infrared was studied in first order. The time-averaged total power output over all lines was measured by a barrier layer cell (FÉSS-U10).

Figure 7a shows the dependence on the gas pressure of the total power of the ultraviolet laser action of nitrogen for the 3-mm diameter tube. One curve was obtained without cooling the tube, and the other with cooling to a temperature close to the temperature of liquid nitrogen. As is seen from the drawing, the optimal pressure of the gas decreases upon cooling. In our experiments, as is usually the case in laboratory setups,

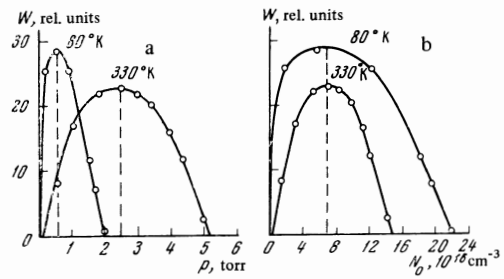


FIG. 7. Output power of ultraviolet laser action of molecular nitrogen: a – as a function of pressure  $p$  and b – density  $N_0$  of the working gas for two temperatures: 80 and  $330^\circ\text{K}$ .

a large ballast gas volume was connected to the discharge tube. This volume remained at room temperature. In this connection, the gas pressure  $p$  in the tube does not change upon cooling, but the density increases like  $N_0 = p/kT$ . Inasmuch as the rate of excitation of the working levels is determined by the population  $N_0$  of the ground state of the molecule, the dependence of the output power on the density has the most importance, and not the dependence on the pressure. Such a dependence is shown in Fig. 7b for the same experimental conditions. As can be seen, the optimal gas pressure remains constant under change in temperature. A similar result is obtained for the He-Ne laser.<sup>[2]</sup>

In Fig. 8, curve 1, the temperature dependence is given for the total power, summed over all the lines, of the ultraviolet laser action, obtained for a constant density of the gas,  $N_0 \approx 6.5 \times 10^{16} \text{ cm}^{-3}$ , corresponding to a maximum power output. When the temperature is reduced to one-third, the power output increases by about 1.5. Direct measurements of the gain as a function of the temperature were not carried out. Such measurements entail serious difficulties, particularly because considerable superradiance is observed even for short active lengths. Qualitatively, however, a strong increase in the gain is observed upon cooling of the gas. In particular, at liquid-nitrogen temperature, in the 3-mm tube, a strong superradiance is observed in the  $0 \rightarrow 0$  band ( $3371 \text{ \AA}$  edge) generally without a mirror, and in the  $0 \rightarrow 1$  band ( $3577 \text{ \AA}$  edge) with a single mirror, while at room temperature, superradiance is observed only on the strong band  $0 \rightarrow 0$  and only with one mirror.

The spectrum of ultraviolet radiation undergoes significant changes. A redistribution of the intensity over the rotational structure takes place. The maximum of the distribution is shifted in the direction of lower  $J$ . Whereas without cooling the maximum power output

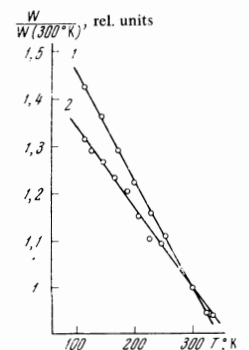


FIG. 8. Relative change in the total output power of ultraviolet (1) and infrared (2) laser action of molecular nitrogen.

corresponds to lines with  $J'_m \approx 8-9$ , at the temperature of liquid nitrogen  $J'_m \approx 5$ . For all the temperatures studied, the output was observed in practice only on the P-branches. Upon cooling, a sharp increase in the intensity of the output is also observed in the band  $0 \rightarrow 1$ , but it is always weak at room temperature. For this band, the power output increases with decrease in the temperature much more rapidly than the total output. Finally, upon cooling, a weak laser action also appears at one band which previously, insofar as we know, it had never been observed. This also indicates a strong increase in the gain. Measurements of long waves have shown that the new band  $1 \rightarrow 0$  (3159 Å edge) also belongs to the second positive system of nitrogen. The rotational structure of the new band is shown in Table I. The spectrum of the output was photographed in first order. The accuracy of measurement of the wavelength was estimated to be 0.03 Å. The calculated frequencies of the lines, given in Table I, were obtained with the use of the data of Coster et al.<sup>[26]</sup> and Dieke and Heath<sup>[27]</sup>.

Some studies of the infrared output on the first posi-

Table I. Rotational structure of laser action on band  $1 \rightarrow 0$  of the second positive system of the nitrogen bands.

$\lambda_{\text{meas}}, \text{Å}$ air	$\lambda_{\text{calc}}, \text{Å}$ air	$\nu_{\text{calc}}, \text{cm}^{-1}$ vacuum	$\Omega$		
			0	1	2
3159.19*	3159.22	31644.6			$P_{39}^c$
	59.02	644.8			$P_{310}^c$
	59.18	645.1			$P_{38}^c$
59.11	59.17	645.2			$P_{311}^c$
	59.12	645.6			$P_{37}^c$
	59.09	645.9			$P_{312}^c$
59.00	59.01	646.7			$P_{36}^c$
	58.98	647.0			$P_{313}^c$
	58.94	647.4			
58.91	58.93	647.5	$P_{210}, 11, 11^c$		
	58.90	647.8	$P_{29}$		
	58.89	647.9	$P_{212}^c$		
58.83*	58.88	648.0	$P_{29}^c$		
	58.81	648.1			$P_{35}^c$
	58.85	648.3			$P_{314}^c$
58.74	58.84	648.4	$P_{28}, 8^c$		
	58.82	648.6	$P_{213}^c, 13$		
	58.76	649.2	$P_{27}$		
58.70	58.74	649.4	$P_{27}^c$		
	58.72	649.7	$P_{214}^c$		
	58.69	649.9			$P_{34}^c$
58.61*	58.65	650.4			$P_{315}^c$
	58.64	650.5	$P_{11}^c, 12$		
	58.62	650.6	$P_{26}^c, 6$		
58.53	58.61	650.7	$P_{11}, 12^c$		
	58.60	650.8	$P_{10}, 13^c$		
	58.59	650.9	$P_{13}$		
58.44	58.58	650.0	$P_{10}^c$		
	58.57	651.1			$P_{215}^c$
	58.54	651.4	$P_{19}^c, 14$		
58.32	58.52	651.6	$P_{19}, 14^c$		
	58.46	652.2			$P_{25}, 5^c$
	58.44	652.4	$P_{18}, 15^c$		
58.27	58.43	652.5	$P_{15}^c$		
	58.42	652.6	$P_{18}^c$		
	58.32	653.7	$P_{17}^c$		
58.16	58.31	653.8	$P_{17}$		
	58.29	654.0			$P_{24}$
	58.27	654.1			$P_{24}^c$
58.03	58.17	655.2	$P_{16}$		
	58.15	655.3	$P_{16}^c$		
	58.04	656.4			$P_{23}^c$
57.98	58.03	656.5			$P_{23}$
	57.99	657.0	$P_{15}^c$		
	57.98	657.1	$P_{15}$		
57.78	57.78	659.0	$P_{14}^c$		
	57.77	659.1	$P_{14}$		
	57.54	661.4	$P_{13}^c$		
57.56	57.54	661.4	$P_{13}$		
	57.52	661.7	$P_{13}^c$		

Note: c - intense  $\Lambda$ -components; \* - most intense lines

tive system of molecular nitrogen,  $B^3\Pi_g \rightarrow A^3\Sigma_u^+$  were also carried out with the 8 mm tube. In Fig. 8, curve 2, the dependence is given for the total output power, over all lines, of the infrared laser action as a function of the temperature, at a constant gas density  $N_0 \approx 1.9 \times 10^{17} \text{ cm}^{-3}$ , which corresponds to the maximum power. As can be seen from the drawing, even in this case a significant increase of the total power is noted upon cooling of the gas. Strong changes take place in the spectrum of the infrared laser action. In addition to the shift in the maximum of the power in the direction of smaller  $J$ , a significant increase is observed in the laser power of certain bands. In particular, the power output in the bands  $4 \rightarrow 2$  ( $\lambda \approx 7500 \text{ Å}$ ) and  $3 \rightarrow 1$  ( $\lambda \approx 7600 \text{ Å}$ ) is observed. At temperatures below  $150^\circ \text{K}$ , the laser action in these bands can easily be observed by eye on white paper in the form of a dark red spot. Appreciable changes are observed also in other bands. More detailed investigations of the infrared laser action of nitrogen upon cooling will be reported elsewhere.

Finally, we note that the preliminary experiments carried out by us, with visible laser action in the transitions of the system of Angstrom bands of the CO molecule<sup>[28,29]</sup> also indicate the appreciable (by as much as an order of magnitude) increase in the laser power upon cooling of the gas to  $\sim 100^\circ \text{K}$ .

A significant increase in the laser power for a series of bands in nitrogen and for the laser action in the case of CO can be connected with the fact that one is operating close to threshold for these transitions at ordinary temperatures. Upon lowering of the temperature, the gain of the medium increases sharply and exceeds the threshold value, which also leads to a strong increase in the power. In this same case, when the gain exceeds the threshold by a large amount even at room temperature, as, for example in the lasing in the strong bands of the first and second positive systems of nitrogen, upon appreciable increase in the gain, only a redistribution of the intensity over the rotational structure is observed and the power increases only slightly. It must also be noted that in our experiments the temperature dependence of the power was measured with the same resonator. At the same time, to get a maximum power for each temperature it is necessary to set the transmission of the mirrors each time at the optimal value. The optimal transmissions as is well known,<sup>[30]</sup> depends strongly on the gain. We did not have the opportunity of carrying out such an optimization.

#### 4. DISCUSSION OF RESULTS

It is of interest to consider the data obtained on the temperature dependence from the viewpoint of the mechanism of production of the inversion in the transitions studied. As regards the mechanism of the production of inversion at the transitions of the second positive system of nitrogen, which correspond to the ultraviolet laser action, there is uniformity of view. All authors assume<sup>[8,13,20,21]</sup> that the dominating mechanism in this case is direct excitation by the electrons from the ground state of the molecule. For such a mechanism, the excitations of the quantities  $N_V'$  and  $N_V''$  should not depend on the gas temperature. The experimental results that we obtained agree well with the calculation

carried out above relative to this case. The character of the change in the lasing spectrum and the significant growth in the gain agree fully with the results of the calculation. However, the accuracy of measurement of  $J'_m$  and the temperature of the working gas used in our experiments did not allow us to distinguish which case of rotational relaxation occurred.

We note further that the discovery of new laser action in the band  $1 \rightarrow 0$  also gives evidence in support of the use of direct excitation by electrons. If we assume that the population of the vibrational levels of the state  $C^3\Pi_u$  is proportional to the corresponding Franck-Condon factor for the transition  $^1\Sigma_g^+(v=0) \rightarrow C^3\Pi_u(v')$ , as is usually the case for direct excitation by electrons, and to assume that the probability of the transition  $C^3\Pi_u \rightarrow B^3\Pi_g$  is also proportional to the corresponding Franck-Condon factor, then the relative value of the gain for  $\lambda \gg 1$  is expressed in terms of the product of these factors, the calculated values of which are shown in Table II for the different bands of the second positive system.

According to this table, the band  $0 \rightarrow 0$  has the greatest gain, then  $0 \rightarrow 1$ , and finally,  $1 \rightarrow 0$ . Just such a sequence has been observed in experiment. Of all the other bands of this system, lasing in which has not yet been observed, the greatest gain is associated with the band  $0 \rightarrow 2$ , the corresponding value for which is also given in Table II.

So far as the infrared emission on transitions of the first positive system of the nitrogen molecules is concerned, there are two points of view as to the mechanism of formation of the inversion in this case. It was assumed in<sup>[20,15]</sup> that even for this system, direct excitation by electrons from the ground state of the molecule plays the dominant role. Mathias and Parker<sup>[10]</sup>, and Bennett<sup>[32]</sup> advanced the opinion that the excitation of the upper working level takes place in smooth fashion as a consequence of the collisions of second order, with energy transfer from the nitrogen molecules to the singlet state  $^1\Pi_g$ . In the first case, in direct excitation by electrons, the behavior of the radiation for cooling of the gas ought to be similar to the behavior of the ultraviolet emission, for which the direct mechanism leaves no doubt.

If the excitation of the working levels involves collisions of second order, then the picture should change, since this process depends strongly on the velocities of the colliding particles.

Inasmuch as the difference in energy between the levels of the state  $^1\Pi_g$ , which is effectively populated by electron excitation, and the levels of the state  $B^3\Pi_g$ , from which emission begins, is sufficiently large, it is necessary to assume that the cross section for energy transfer by second order collisions must decrease rapidly with decreasing temperature. Therefore it can be expected that in the excitation by second order colli-

sions the infrared laser action will fall off with cooling, as is the case for the He-Ne laser,<sup>[1-4]</sup> where the energy transfer by collisions of the second kind serves as the basic process for inversion. However, experiments have shown that the infrared laser action upon cooling behaves in the same fashion as the ultraviolet. Its power and gain are increased. This serves as still more evidence in support of the direct mechanism of inversion in this case, too.

## 5. CONCLUSION

The theoretical calculation given above, as well as the experimental results, show that the cooling of the working gas leads to a significant increase in the gain of the active medium in lasers in electronic transitions in diatomic molecules. The increase in the gain is especially significant for those transitions for which the gain is small. For these same transitions, an appreciable increase in the laser power is observed. In this connection, the cooling of the working gas in molecular lasers can, above all, serve as a method of finding new emission or as the method for the increase of the laser power for those cases in which the system operates close to the laser threshold. An example of such an application is the discovery of a new band of ultraviolet laser action of nitrogen, described in the present work.

The results of the present work show further that in the study of the dependences of the characteristics of the molecular lasers (for example, the output power) on such parameters as the discharge current, the repetition frequency of the pulses, length of pulses and so forth, it is necessary to take into account the change in the temperature of the working gas. In particular, it was found that upon increase in the pulse repetition frequency, the peak power output decreases. Account of heating shows that for low frequencies of repetition, this effect is on the whole determined by the heating of the gas. A detailed account of the temperature change is necessary in the study of the dependence on all these parameters which can affect the temperature of the working gas.

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Table II

Band	Edge of band, Å	Product of the Franck-Condon Factors <sup>[31]</sup>
0→0	3371	0.255
0→1	3577	0.185
1→0	3159	0.118
0→2	3805	0.081

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