Generation in the 3.3–3.6-μm region in condensed nitrogen

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(Submitted 7 April 1979)

Pis'ma Zh. Eksp. Teor. Fiz. **29**, No. 11, 688–692 (5 June 1979)

High-power generation is obtained at electronic transitions $\omega \rightarrow a$ and $a \rightarrow a'$ in a deeply-condensed N_2 molecule in an electric-discharge laser with transverse excitation. Possible mechanisms of inversion at these transitions are first discussed, as are reasons for a sharp increase in the output power in the case of cooling.

PACS numbers: 42.55.Mv

The 3-μm wavelength region is highly interesting for studying the resonant interaction of laser radiation with matter. One possible way of obtaining generation in this region is to use the electronic transition $\omega^1\Delta_u \rightarrow a^1\Pi_g$ (0–0 vibrational band) in a N_2 molecule in which a weak laser action was observed by McFarlane as early as 1966.[1,2] The nitrogen molecules were excited by a pulsed discharge along the laser tube axis at the room temperature. Because this system of generating laser radiation remained unnoticed at that time, McFarlane's attention was focused on the spectroscopic investigations and he failed to explain the mechanism for producing an inversion. Earlier, while using the same experimental setup, McFarlane obtained laser action in molecular nitrogen due to another singlet, $a^1\Pi_g \rightarrow a'^1\Sigma_u^-$.[3,4]

We obtained concurrent pulsed generation at the N_2 electronic transitions $\omega \rightarrow a$ and $a \rightarrow a'$, emitting in the 3.6- and 3.3-μm regions, respectively. Upon cooling the N_2-He mixture to the liquid nitrogen temperature; we obtained ~0.5-mJ energy in a 0.5-μsec pulse.

The experimental setup consisted of a laser chamber containing liquid-nitrogen-cooled profiled aluminum electrodes each 950 mm long and 25 mm wide. The interelectrode spacing was 25 mm. The working gas was excited by a discharge of a 2×10^{-2}-μF capacitor at an initial voltage of 30 kV. The shape of the discharge current was a damped sinusoidal with the initial period of 330 nsec and the current amplitude at the first maximum was up to 2 kA. The working gas consisted of an 1 : 14 N_2-He mixture at the total pressure of 20 tor. The resonator consisted of two Au-coated mirrors with $R_1 = 5$ m and $R_2 = \infty$. The output from the cavity was coupled out by means of a beam splitter.

Strong laser action was observed at five lines in the 2–1 band of the $a^1\Pi_g \rightarrow a'^1\Sigma_u^-$ system and the 0–0 band of the $\omega^1\Delta_u \rightarrow a^1\Pi_g$ systems. The total output power due to all the lines attained 1 kW.

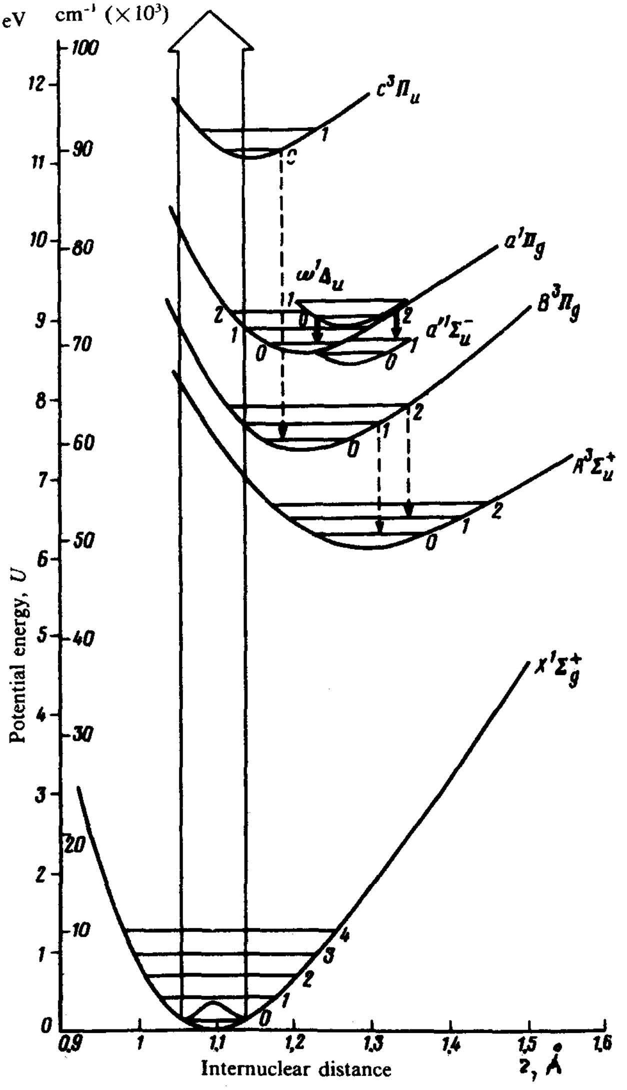

FIG. 1. Energy level diagram of N_2 molecule.

Figure 1 shows an energy level diagram for an N_2 molecule. It shows the potential curves of the electronic states with several lower vibrational levels. In addition to the ground state $X\,^1\Sigma_g^+$ and the states $\omega^1\Delta_u$, $a^1\Pi_g$ and $a'^1\Sigma_u^-$ taking part in the laser action, the triplet states $C\,^3\Pi_u$, $B\,^3\Pi_g$ and $A\,^3\Sigma_u^+$ are also shown. As is known,[5] strong pulsed generation due to the $C\,^3\Pi_u \rightarrow B\,^3\Pi_g$ and $B\,^3\Pi_g \rightarrow A\,^3\Sigma_u^+$ transitions at 0.34 and 0.9 μm, respectively, is in evidence. The population inversion mechanism for the above is excitation of the upper laser levels by a direct electron shock in accordance with the Frank-Condon principle. The broad vertical arrow pointing up (Fig. 1) indicates excitation by an electron shock. The dotted arrows indicate the strongest laser transition in the triplet states. Moreover, the pumping and emission channels which are always coupled to the turning points of the vibrational levels, are noticeably spaced.

The foregoing mechanism of population inversion is common to all well-studied systems based on bound-bound electronic transitions for both N_2 and other molecules, and it could occur in the $a^1\Pi_g \to a'^1\Sigma_u^-$ system. In contrast to the $a \to a'$ system, generation due to vibrational transition in the 0–0 system $\omega \to a$ may not be readily described by the foregoing mechanism. The upper laser state is considerably displaced with respect to the molecular ground state and the vibrational level $v' = 0$ is located at a considerable distance from the electron excitation channel. In addition to this, the pumping transition $X^1\Sigma_g^+ \to \omega^1\Delta_u$ is strongly forbidden ($\Delta\Lambda = 2$), which violates the selection rule with respect to projection of the orbital angular moment $\Delta\Lambda = 0, \pm 1$. Also of consequence is the fact that we observed in this system a delay (approximately 1 μsec at 20 tor) between the pumping and laser pulses. The magnitude of the delay was inversely proportional to pressure. It should be pointed out that a delay was also observed for the 2→1 transition in the $a \to a'$ system, which to a certain degree also casts doubt on the direct excitation mechanism for this system.

Thus, the heretofore existing mechanism considered unique in gas-discharge lasers for producing a population inversion due to electronic transitions between the bound molecular states by means of a direct electron shock is not a result of transitions in the 2–1 system $a \to a'$ at the 0–0 system $\omega \to a$. Most probably, the inversion is due to a recombination processes or radiative cascading. One such cascade may be the known transition $y^1\Pi_g \to \omega^1\Delta_u$ (the second Kaplan system).

Since a sufficient thermalization of excited nitrogen states occurs during the period of generation, it appears possible to substantially increase the gain at the individual rotational transitions by decreasing the gas temperature.[6] An expression for the gain at the center of a Doppler-broadened electron-vibrational-rotational transition $n'v'J' \to n''v''J''$ in the case of a totally depopulated lower level is

$$K(\nu_0) = \frac{hc^4\sqrt{M}}{8\pi k\sqrt{2\pi kN}} \, \frac{g_s}{g_s^s + g_s^a} \, \frac{A_{n'n''} q_{v'v''} N'_v B'_v}{\nu_0^3 T\sqrt{T}} \, S_{J'J''} \exp\left(-\frac{F(J')}{kT}\right),$$

where ν_0 is frequency at the line center; $A_{n'n''}$ is the probability of an electron transition; $q_{v'v''}$ is the Frank-Condon coefficient of the vibrational transition $v' \to v''$; $S_{J'J''}$ is the Henle-London coefficient for the rotational transition $J' \to J''$; $F(J') = B'_v J'(J'+1) - D'_v[J'(J'+1)]^2$ is energy of the rotational term of the upper state with the quantum number J'; N'_v, B'_v and D'_v is the population and the rotational constants of the upper vibrational level with the quantum number v'; g_s is the nuclear statistical weight of either the symmetrical (g_s^s) or antisymmetrical (g_s^a) states depending on where the observed line originates; M is molecular weight in atomic units; N is Avogadro's number; c is the speed of light; and h and k are Planck's and Boltzmann's constants, respectively.

Clearly, a strong inverse dependence of gain on the gas temperature is in evidence. This is highly apparent from Fig. 2 which shows the calculated dependence of gain in the P, Q and R branches in the 0–0 band of the $\omega \to a$ transition on the rotation-

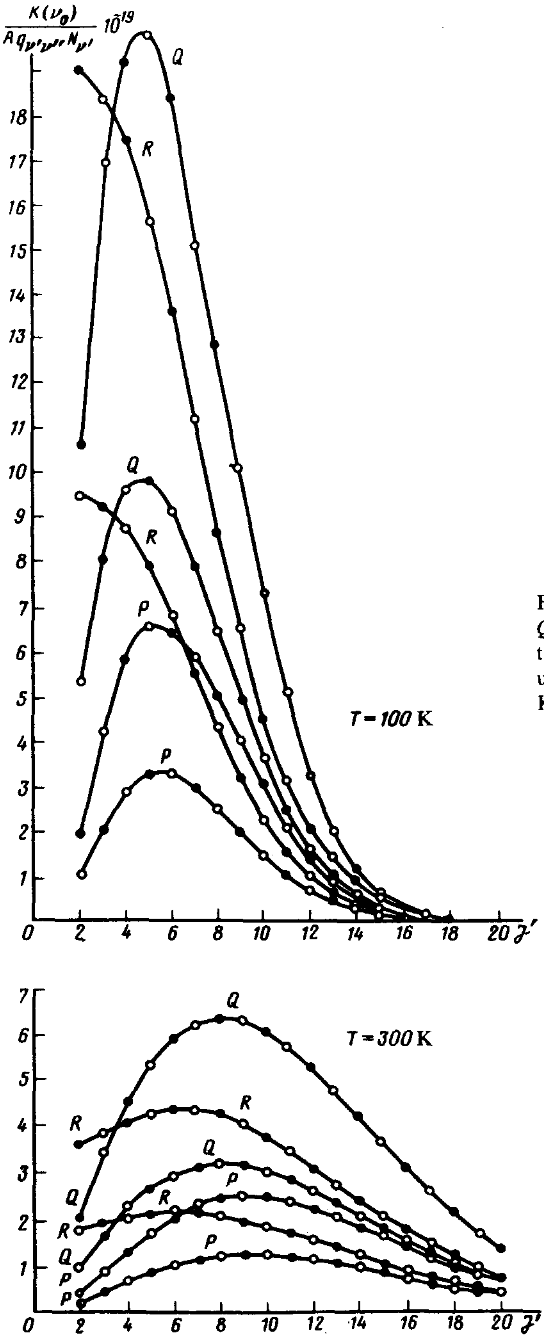

FIG. 2. Calculated dependence of gain in the P, Q and R branches of the 0–0 band $\omega \rightarrow a$ transitions on rotational quantum number J' of the upper level for two temperatures, 300 and 100 K.

al quantum number J' of the upper level for two temperatures 300 and 100 K. In view of each rotational level in both electronic states being split into two very close sublevels (Λ-twinning), each of the P, Q and R branches consist of two series of lines which form closely-coupled doublets. In the figure, these series are shown as dark (*d*-series) and light (*c*-series) circles. Intensity rotation occurring in each series is associated with the presence in a gas of the ortho- and para-phases of nitrogen.

Thus, we showed that the use of a laser operating at the 2–1 transitions of the $a^1\Pi_g \rightarrow a'^1\Sigma_u^-$ system and the 0–0 transitions of the $\omega^1\Delta_u \rightarrow a^1\Pi_g$ system of N_2 molecule is promising for obtaining relatively powerful radiation in the important 3.3- and 3.6-μm regions under the conditions of deep cooling and transverse discharge of a large volume of gas. It is assumed that a new cascade mechanism for producing the population inversion is in evidence in these N_2 systems.

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