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Study of the UV emission of an inductive nitrogen laser*

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***Abstract*. Lasing at 337.1 and 357.7 nm is obtained upon excitation of nitrogen molecules by a toroidal pulsed inductive discharge. A system for formation of a pulsed inductive discharge for exciting gas lasers is described. The spontaneous emission spectra of the nitrogen inductive discharge plasma and the emission spectrum of the nitrogen laser are recorded and interpreted. The dependences of the energy and duration of laser pulses on the resonator Q factor and pump level are studied. The output energy of the inductive nitrogen laser emitting 15 ± 1-ns pulses achieves 4.5 mJ. The generation of high-power 300-kW pulses is obtained for the first time at a low ($\sim$ 1 Torr) pressure of pure nitrogen. The spatial distribution of the laser radiation intensity in the discharge tube cross section is investigated. The cross section of the radiation beam of the inductive nitrogen laser had the shape of a ring with the external diameter of 34 mm and width $\sim$ 4 mm, its divergence being $\sim$ 0.8 mrad. The average output power of the laser achieved 120 mW at a pulse repetition rate of 30 Hz.**

Keywords: *pulsed inductive discharge, nitrogen laser*.

1. Introduction

The fabrication of a pulsed inductive UV molecular nitrogen laser was first reported in 2007 [1]. By exciting molecular nitrogen by a toroidal pulsed inductive discharge, 2.4 mJ of output lasing power was achieved at 337.1 nm at a pressure of 0.6 Torr.

The aim of this paper is to show that a pulsed inductive discharge is an efficient alternative tool for exciting gas lasers, offering a number of advantages compared to transverse and longitudinal electric discharges. We present here the results of experimental studies of spectral, temporal and energy parameters of the UV radiation of an inductive nitrogen laser. The main task was to study the emission properties of molecular nitrogen in a pulsed inductive discharge and to increase the output energy of the laser.

By a pulsed inductive discharge is meant in this case the inductively coupled plasma produced by vortex electric currents due to electromagnetic induction created by an alternating magnetic field in a gas medium. The inductive discharge differs from the longitudinal and transverse electric discharges in that it is produced in gas without using electrodes.

The nitrogen laser attracts interest because of wide applications of its UV radiation at 337.1 nm. This laser is one of the most popular high-power UV radiation sources used for pumping dye [2] and semiconductor [3] lasers, in spectroscopy, photochemistry [4] and medicine [5], as well as in diagnostics.

Lasing in molecular nitrogen at 337.1 nm is usually obtained by exciting nitrogen and its mixtures with other gases with the help of longitudinal and transverse pulsed electric discharges [6]. Although upon excitation of small active volumes (up to 10 cm^3), the output energy is low (0.1 – 0.2 mJ), it is possible to fabricate lasers with high pulse repetition rates, up to 10 kHz and above [7]. By using a transverse discharge to excite a nitrogen laser, it is possible to increase the active volume up to 30 – 100 cm^3. The output energy of such lasers is considerably higher and can achieve 10 mJ [8] and more [9]. The operating pressure of pure nitrogen in these lasers is 20 – 50 Torr, while in the mixtures of nitrogen with other gases (He, Ar, SF_6, NF_3, etc.) this pressure is from 100 Torr to a few atmospheres. This leads to technical difficulties in the development of excitation systems that should provide a homogeneous volume discharge in nitrogen used as the active medium of the laser. The parameters of the discharge strongly affect the output power and duration of laser pulses, the lasing stability and the operating life of the gas medium. As a result, the pulse repetition rate in a transverse discharge does not exceed a few tens of hertz. Because pulsed electric discharges are produced between metal electrodes, the material of electrodes is sputtered during laser action, resulting in the formation of cathode spots and streamers and, finally, in quenching of lasing. Thus, it is necessary to replace not only the working gas but also electrodes and optical elements of the laser resonator.

Because a pulsed inductive discharge is produced without using any electrodes, it is free from the above-mentioned problems restricting applications of the laser. In addition,

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the pulsed inductive discharge, which represents the inductively coupled plasma produced by vortex currents, is of interest itself as a tool for creating the active laser medium and achieving inversion at atomic and molecular transitions. The formation mechanism of this discharge is of interest because in this case there are no certain concepts about the parameter E/p ($E = U/d$, where U is the voltage applied to electrodes between which the discharge is produced; d is the distance between electrodes; and p is the pressure of nitrogen [10], which plays a key role in the development of electric-discharge nitrogen laser. Therefore, the investigation of pulsed UV inductive nitrogen lasers is of current interest.

The properties of the UV nitrogen laser have been studied in many papers (see, review [6]). The inversion formation mechanism at the $\mathrm{C}\,^3\Pi_\mathrm{u} \rightarrow \mathrm{B}\,^3\Pi_\mathrm{g}$ transition in molecular nitrogen is well studied and described in [11, 12]. It involves the direct electron-impact excitation of molecules from the $\mathrm{X}\,^1\Sigma_\mathrm{g}^+$ ground state during which the predominant population of the upper working levels occurs in accordance with the Franck–Condon principle. The radiative lifetime τ_C of a nitrogen molecule on the $\mathrm{C}\,^3\Pi_\mathrm{u}$ upper laser level is ~ 38 ns [13] ($\tau_\mathrm{C} \ll \tau_\mathrm{B}$) and is determined mainly by the transition to the $\mathrm{B}\,^3\Pi_\mathrm{g}$ lower laser level with the lifetime $\tau_\mathrm{B} \approx 9$ μs [13]. The radiative transition from the $\mathrm{B}\,^3\Pi_\mathrm{g}$ level can occur only on the metastable $\mathrm{A}\,^3\Sigma_\mathrm{u}^+$ level. Therefore, under normal conditions, the depopulation rate of the $\mathrm{B}\,^3\Pi_\mathrm{g}$ level due to collisions of nitrogen molecules with other particles is comparatively small, and stationary inversion cannot be achieved at the $\mathrm{C}\,^3\Pi_\mathrm{u} \rightarrow \mathrm{B}\,^3\Pi_\mathrm{g}$ transition. The inversion can be obtained only in the nonstationary regime at the excitation pulse front under the condition that the excitation rate of the upper level is higher than that of the lower level. Such transitions are called self-contained transitions. To produce a homogeneous volume electric discharge and obtain lasing at the transitions of the second positive band system of molecular nitrogen in nitrogen lasers described in the literature, the condition $E/p \geqslant 200$ V cm^{-1} Torr^{-1} should be fulfilled. Because electrodes are absent in the case of the inductive discharge, the parameter E/p becomes uncertain. However, the general concept of UV nitrogen lasers was assumed invariable, and it was necessary to produce the inductively coupled plasma with electronic parameters providing the inversion mechanism described above, namely, to obtain electrons in plasma with concentration $10^{14} - 10^{15}$ cm^{-3} and energy 12–16 eV [14].

2. Experimental setup

A cylindrical inductive toroidal discharge in nitrogen was excited in our experiments by using an electrical circuit presented schematically in Fig. 1. This excitation system differed from that described in our paper [1] and provided more efficient energy transfer from a storage capacitor C_1 to the active medium. To increase the voltage across an inductor and increase the energy input into gas, an additional capacitor C_3 was used in the circuit. The excitation system operated in the following way. The capacitor C_1 (80 nF) was charged from an ALE 152A Lambda EMI pulsed power supply up to the voltage 20–27 kV of the positive polarity. In this case, the energy of 16–30 J was stored in the capacitor. When the voltage across capacitor C_1 achieved the maximum, a triggering pulse was fed to a high-voltage switch (TPI 10k/20 thyratron). After the thyratron actuation, the capacitor C_1 began to discharge, a negative voltage appeared across choke L_1, and energy was transferred to capacitors C_2 and C_3. Both capacitors were charged during the time 1.5–2.0 μs up to the breakdown voltage of a spark gap. The capacitor C_3 and the spark gap represented a low-inductive circuit in which the capacitor C_3 began to discharge after the actuation of the spark gap slightly earlier than the capacitor C_2. During the discharge of the latter, an alternating current flowed through an inductor L_2, which produced an inductive discharge in the discharge tube. The ignition instant of the inductive discharge was determined by the appearance of spontaneous emission of nitrogen in the discharge tube.

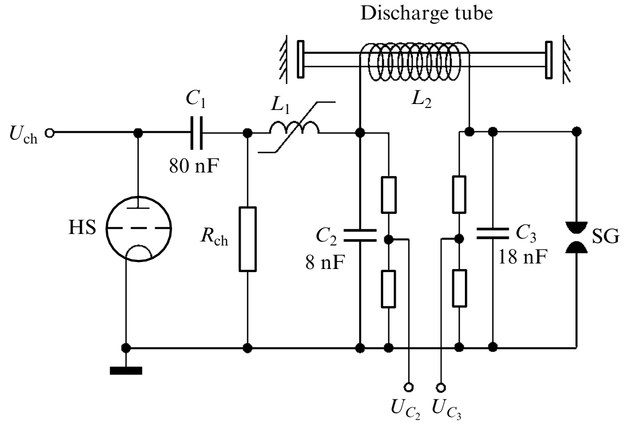

Figure 1. Electric circuit of the excitation system of a N_2 laser by a pulsed inductive cylindrical discharge: (HS) high-voltage switch (TPI1-10k/20 thyratron); (SG) uncontrollable gas-filled spark gap; (L_1) charging choke; (L_2) inductor.

The spontaneous emission spectra of the inductive discharge in nitrogen and the lasing spectrum were recorded with a S-150 Solar LS spectrometer with a resolution of 0.66 nm in the spectral range from 200 to 1100 nm and a SpectraPro-500 Acton Research Corp. spectrograph with a resolution of 0.025 nm in the spectral range from 180 to 700 nm. The output laser energy was measured with a PE50-BB Ophir pyroelectric detector. The temporal parameters of electric pulses were recorded with high-voltage P6015A probes and a 200-MHz TDS-2024 Tektronix oscilloscope. The temporal parameters of optical pulses were recorded with a FEK-22 coaxial photocell with a resolution of 10^{-10} s. To avoid distortions of the shape of pulses by a photodetector, the radiation intensity was controlled with the help of UFS-1, UFS-6, etc. filters. The spatial distribution of the laser radiation intensity over the tube cross section and the light beam profile were analysed by using a WinCamD-UCM digital video camera (Data Ray Inc.).

It was found in [1] that the output energy of the inductive nitrogen laser was proportional to the discharge tube diameter, i.e. the lasing efficiency and output energy increased with increasing the tube diameter. Because of this, we used in our experiments a ceramic discharge tube with inner diameter of 34 mm and external diameter of 40 mm. The tube was sealed by means of plane-parallel quartz windows oriented perpendicular or at the Brewster angle to the tube axis (Fig. 1). The optical resonator was formed by external plane dielectric mirrors. The rear mirror had the reflectance $R_1 = 99\,\%$ in the spectral region 300 – 380 nm. The reflectance of the output mirror was optimised during experiments to obtain the maximum output energy. Mirrors with reflectances from 8 % to 93 % were used.

The inductor L_2 consisted of separate sections representing solenoids made of a cable wire of cross section 4 mm^2. The results presented in this paper were obtained by using the inductor containing 20 sections, each of them consisting of five coils. The solenoids were connected in parallel, and the total length ($\sim$ 60 cm) of the inductor was assumed the length of the active medium of the nitrogen laser. Nitrogen or its mixtures with other gases were admitted from a gas system into the tube up to pressures 0.1 – 10 Torr. Gases flowed longitudinally during experiments.

3. Results and discussion

Experiments with the inductive discharge in pure nitrogen showed that the pressure range in which the inductively coupled plasma can exist is quite narrow, from 0.1 to 10 Torr. Visual observations with the use of different optical filters showed that the inductive discharge was homogeneous in the entire pressure range. No sparks and streamers were observed in the inductive discharge in pure nitrogen and its mixtures. Judging from the obtained oscillograms of voltage across capacitors C_2 and C_3, the energy input to the inductively coupled plasma decreased with increasing the nitrogen pressure. At minimal nitrogen pressures (lower than 0.1 Torr), the inductive discharge almost completely filled the discharge tube over the entire inductor length and the tube cross section. As the nitrogen pressure was further increased (above 0.2 Torr), the emission intensity of the inductive discharge in the central part of the tube began to decrease rapidly. The discharge took a toroidal shape and concentrated near the inner wall of the tube. The discharge was observed in the form of a luminous cylinder with the walls of thickness decreasing with increasing pressure. At nitrogen pressures exceeding 4 Torr, the discharge emission intensity decreased, and at a pressure of $\sim$ 10 Torr the inductive discharge abruptly terminated. At the inner surface of the discharge tube, a weakly emitting electric discharge was detected which we considered as a capacitive discharge. The spontaneous emission spectrum of the pulsed inductive discharge plasma in nitrogen was studied in the absence of resonator mirrors with sealing windows oriented at an angle to the tube axis close to the Brewster angle. The spectrum shows that all the emission lines are concentrated in the second positive system of bands of molecular nitrogen in the spectral range from 300 to 400 nm (Fig. 2a). The ratio of band intensities in the spectrum depended on the nitrogen pressure. At pressures 0.2 – 3.0 Torr, the two most intense lines were observed at 337.1 nm (the 0 – 0 transition) and 357.7 nm (the 0 – 1 transition).

After the mounting of dielectric mirrors and alignment of the optical resonator, we obtained UV lasing at transitions of molecular nitrogen excited by the pulsed inductive discharge in the pressure range from 0.3 to 3 Torr. The maximum of the lasing efficiency was achieved at pressures 0.5 – 0.8 Torr. Investigations of the vibrational structure of the emission spectrum of the nitrogen laser showed that, unlike the results obtained in [1], lasing was observed not at one but at two lines corresponding to two most intense spontaneous emission lines at 337.1 nm (the 0 – 0 transition) and 357.7 nm (the 0 – 1 transition) (Fig. 2b). Unlike the spontaneous spectrum, the intensity ratio for these lines proved to be rather high. The intensity of the 337.1-nm line exceeded that of the 357.7-nm line more than by two orders of magnitude. It is interesting that simultaneous lasing at these two lines in pure nitrogen at low pressures (1 – 2 Torr) was previously reported only in one paper [15]. In experiments [15], nitrogen was excited by a longitudinal electric discharge of duration 1.5 μs in a tube of length 90 cm with the inner diameter 3 mm. Only the laser emission spectrum was studied, and the energy and temporal parameters of laser emission were not reported. The results obtained in [15] proved to be useful for our investigations and we used them to interpret the rotational structure of the emission spectra of the inductive nitrogen laser in the (0 – 0) and (0 – 1) bands because we have failed to record the detailed rotational structure of these lines due to a low spectral resolution of our spectrometers. The widths of the 337.1-nm and 357.7-nm lines were measured to be 4.5 Å. According to [15], we found that the laser emission spectrum in the (0 – 0) band contained 32 lines related to 46 rotational transitions (mainly in the P branch) with wavelengths from 336.6912 to 337.1437 nm. Emission in the (0-1) band is caused by the 12 rotational transitions of the P band and consists of four lines with wavelengths from 357.6112 to 357.695 nm.

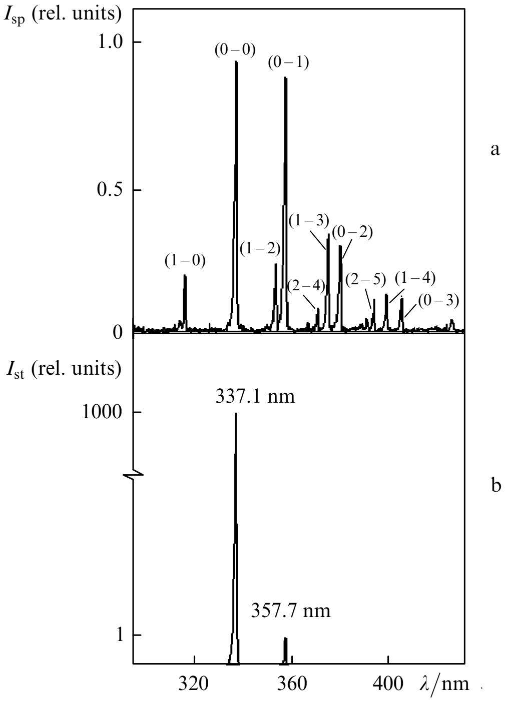

Figure 2. Spontaneous emission spectrum of the inductive discharge in nitrogen (a) and the emission spectrum of the inductive nitrogen laser (b); $U_{ch} = +25$ kV.

We studied the temporal dependences of excitation, spontaneous and inductive nitrogen laser pulses. Figure 3 shows the oscillograms U_{C_2} and U_{C_3} of voltage pulses across capacitors C_2 and C_3, respectively. These oscillograms were compared with oscillograms of spontaneous UV radiation pulses I_{sp} of the inductive discharge in nitrogen and laser pulses I_{st}. One can see that, unlike electric-discharge nitrogen lasers, the onset of pulsed lasing I_{st} does not coincide in time with the onset of the spontaneous emission pulse I_{sp}. Lasing appears and proceeds during the time period corresponding to the maximum voltage gradient across the capacitor C_2. The spontaneous emission I_{sp} also appears during this time period, but 60 – 70 ns earlier than lasing and its duration is 3 – 4 times longer. By analysing the obtained oscillograms, we concluded that the first small maximum in the spontaneous emission oscillogram I_{sp} is explained by the appearance of a capacitive discharge in nitrogen at the inner surface of the tube. After 50 – 60 ns, the inductive discharge was ignited and the main spontaneous UV emission peak (with the maximum amplitude and duration) of nitrogen appeared simultaneously, which corresponded to the appearance and development of the inductive discharge in the discharge tube. In our opinion, the oscillogram of the main spontaneous emission peak I_{sp} reflects the shape of the inductive discharge current pulse in nitrogen. The inductive and capacitive discharges further developed simultaneously in time until the end of the inductive discharge. The capacitive discharge existed during the entire recharging time of the capacitor C_2 and, therefore, during the existence of alternating current in the inductor L_2.

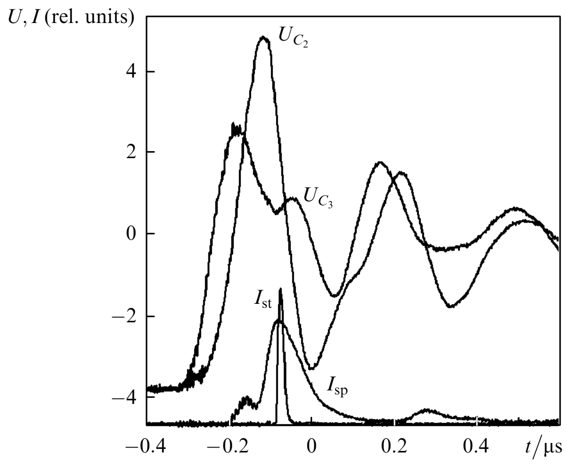

Figure 3. Oscillograms of voltage pulses across capacitors C_2 and C_3 and spontaneous emission pulses I_{sp} of the inductive discharge plasma in nitrogen and nitrogen laser pulses I_{st}; $U_{ch} = +25$ kV.

To increase the output energy of the inductive nitrogen laser compared to that obtained in [1], we optimised the resonator Q factor. The results of this optimisation are presented in Fig. 4. This figure shows that the maximum output energy (4.5 mJ) is achieved for $R_2 = 60\,\%$. The laser pulse FWHM was 15 ± 1 ns. As the reflectance of the output mirror was further increased, the output energy decreased. However, the laser pulse duration increased from 13 ± 1 ns for $R_2 = 16\,\%$ to 18 ± 1 ns for $R_2 = 93\,\%$. This is larger than in electric-discharge lasers, where the pulse duration does not exceed 5 – 15 ns [8, 9]. The duration of UV laser pulses (at the base level) in the dense resonator exceeded 55 ns.

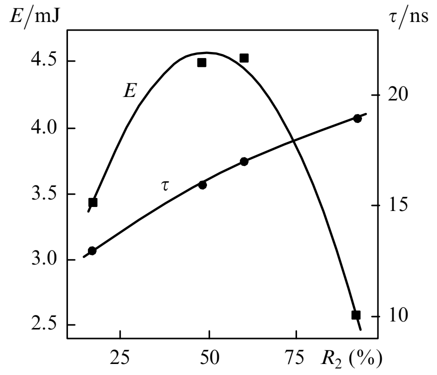

Figure 4. Dependences of the output energy E and the pulse FWHM τ of the inductive nitrogen laser on the reflectance R_2 of the output mirror; the reflectance of the rear mirror is $R_1 = 99\,\%$, $U_{ch} = +27$ kV.

The attempts to use some additions to nitrogen for increasing the duration and power of nitrogen laser pulses and obtaining lasing at the other bands of the second positive system or other band systems gave no results. By adding inert gases He, Ne, and Ar and halogen-containing gases F_2, NF_3, SF_6, CF_6Cl_2 to nitrogen, we found that at their low concentrations (1 % – 10 % of the nitrogen concentration), no changes in the temporal, energy and spectral parameters of laser radiation occurred. At high concentrations of these gases, the output energy decreased in all cases. We can assume that because the active medium in the inductive laser is not connected in series with other elements of the discharge electronic circuit, the addition of other gases to nitrogen cannot affect the operation of the excitation system and formation conditions of the inductive discharge. It can only affect kinetic processes related to excitation and quenching of different levels of nitrogen molecules and to absorption of radiation at different transitions. However, the influence of additions at low total pressures is probably reduced to the collision quenching of the upper laser level and inversion destruction. For this reason, all the results presented in this paper were obtained upon excitation of pure nitrogen by the inductive discharge.

Figure 5 shows the dependence of the output energy E of the inductive nitrogen laser on the charging voltage U_{ch}. One can see that lasing appears at voltages exceeding 20 kV. As the charging voltage is increased, the output energy increases, but not proportionally to the voltage. At voltages above 24 kV, the energy increase rate slowed down. During the measurements of the output energy at different charging voltages, the nitrogen pressure was optimised for each voltage. As the charging voltage was increased from 20 to 27 kV, the optimal pressure increased from 0.5 to

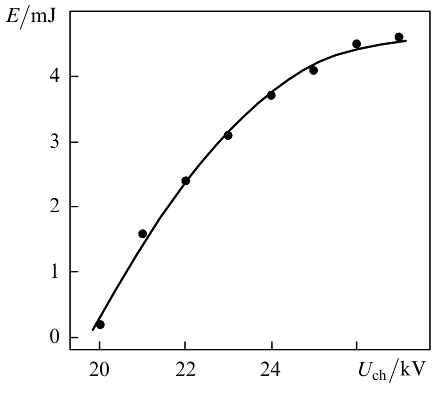

Figure 5. Dependence of the output power E of the inductive nitrogen laser on the charging voltage U_{ch}; $\tau = 15 \pm 1$ ns, $p = 0.5 - 0.8$ Torr.

0.8 Torr. The maximum energy of 4.5 mJ was obtained at the maximum voltage of 27 kV. For 15-ns pulses, this corresponds to the laser pulse power of 300 kW. It is important to note that such a high pulsed power was never achieved before in a nitrogen laser at low pressures ($\sim$ 1 Torr). This result demonstrates the specific features of the operation of the inductive nitrogen laser such as its emission spectrum containing many vibrational lines, which is obtained upon such excitation, and a long duration of pulses with a comparatively flat leading edge (about 7 ns). The total lasing efficiency, obtained as the ratio of the output energy E at the maximum voltage to the energy stored in the capacitor C_1, was about 0.02%. According to our measurements, the pulse-to-pulse instability of the lasing amplitude was ± 1 %.

By using a digital video camera, we studied the spatial distribution of the laser radiation intensity at the discharge tube output. The light-beam cross section had the shape of a ring of external diameter $\sim$ 34 mm. The study of the beam profile showed that the laser radiation intensity at the external boundary of the ring was minimal. The lasing intensity increased towards the ring centre and achieved the maximum at a distance of about 1 mm from the external boundary. Then, the radiation intensity decreased almost to zero at a distance of 4 mm from the boundary. This value can be treated as the ring width. The laser radiation divergence, which was determined by measuring the size of the laser radiation ring at different distances from the laser (0.1 – 10 m), was $\sim$ 0.8 mrad. The circular structure of the beam cross section is a specific feature of pulsed inductive lasers with a cylindrical inductive discharge. Such beams in the case of a low radiation divergence offer certain advantages because they can be focused to produce the radiation intensity distribution similarly to Bessel beams.

We performed experiments with the inductive nitrogen laser operating in the repetitively pulsed regime. The pulse repetition rate was varied from 1 to 30 Hz. We found that the average output power increased linearly with increasing pulse repetition rate. For the repetition rate of 30 Hz and output power of the nitrogen laser of 4 mJ, the average output power was 120 mW. The output energy was independent of the pulse repetition rate because the active medium was cooled at the ceramic tube wall directly adjacent to the lasing region.

4. Conclusions

Our investigations of the pulsed UV inductive laser have revealed a number of specific features caused by differences in the electronic parameters of the inductively coupled plasma and plasma of the transverse and longitudinal electric discharges. One of these features is simultaneous lasing at 337.1 and 357.7 nm in pure nitrogen. The high pulsed laser power (300 kW) and energy (4.5 mJ) have been obtained for the first time at a low nitrogen pressure (about 1 Torr). A long duration of laser pulses (FWHM of 18 ns) with a comparatively flat leading edge (about 7 ns) demonstrates the difference of the inductive discharge from the electric discharge from the point of view of excitation of the laser levels of molecular nitrogen. Note that the laser spectrum observed upon such excitation exhibits many rotational lines and its shape and lasing power and duration are independent of additions to nitrogen. The laser-beam cross section has a characteristic ring shape. The radiation of the inductive nitrogen laser has a low divergence (0.8 mrad) and its pulse-to-pulse instability is within ± 1 %.

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