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The Dependence of the Luminous Efficacy of FPS and SPS Lines of Nitrogen in the Mixture CO_2:N_2: He = 1:1:8 on Acoustoplasma Discharge Parameters and the Selective Amplification of Individual Lines

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Abstract: The results of experimental study of the dependence of the light efficiency of the First and Second Positive Systems of nitrogen lines (FPS and SPS) on the parameters of the acoustoplasmic discharge are presented. The spectral regions FPS (575 - 687nm) and SPS (350 - 435nm) are considered. The working mixture of CO_2 laser (CO_2:N_2:He=1:1:8) was investigated. It is noted that the main portion of the energy in the visible spectrum for such a mixture is concentrated in FPS and SPS nitrogen lines. The data on the selective strengthening of individual spectral lines of nitrogen, depending on the discharge parameters, are presented. Pressures in the region from 0.1 to 50 torr were investigated. The DC discharge is compared with acoustoplasma discharge. The change in the intensity of the strong red line (about 655 nm) from the modulation frequency of the discharge current in the acoustoplasmic mode is given. It has a resonant character and can be changed almost 5 or more times. The shift of the strong red spectral line in pure nitrogen plasma relative to tabular spectral lines was found.

Keywords: SPS and FPS lines of nitrogen, luminous efficacy, acoustoplasma discharge parameters.

1. Introduction

To determine the characteristics of equilibrium plasma, it is enough to know its thermodynamic parameters: temperature and pressure and use the well-known Boltzmann, Maxwell distributions and the Saha equation. The simplest state of nonequilibrium plasma is described by two-temperature model. For detailed description of nonequilibrium molecular plasma, it is necessary to take into account several temperatures (rotational and vibrational for the electrons and for the gas). Sometimes it is generally impossible to characterize the system by any temperature [1,2].

If a pulsed electric field is applied to the initially equilibrium plasma, then the degree of ionization, as a rule, lags behind the temperature increase. For sufficiently long pulses, if the radiation output from the considered plasma volume and the removal of charged particles are weak, then a two-temperature plasma model is realized. If one of these factors has a significant intensity (for example, gas pumping or a narrow discharge capillary), then even for a constantly applied electric field, an "under-ionized plasma" arises in which all three distributions, Boltzmann, Maxwell, and Saha, are not applicable [1].

With decreasing electric field, another group of nonequilibrium states appears, where the ionization decreases more slowly than the temperature. In this case recombining plasma occurs.

Due to the self-consistency of processes in the plasma, the changes in different degrees of freedom are interconnected and the deviation from equilibrium in one of the degrees of freedom can lead to deviation from equilibrium in other degrees of freedom.

In addition, in electric discharge plasma, the multilevel energy structure of atoms and molecules causes great difficulties. In fact, in a nonequilibrium plasma there are many mutually transforming neutral components that differ in process cross sections, ionization energy, etc. The same is the case for ions in nonequilibrium plasma.

In the acoustoplasma (AP) mode of operation of a gas discharge tube, the discharge current has constant and alternating components [3, 4]. The amplitude of the variable component, as a rule, does not exceed the value of the constant component and thus, there is no re-ignition of the discharge, it always exists. But at any given time, the current and voltage at the discharge vary in magnitude from minimum to maximum values and vice versa, i.e., in accordance with the foregoing, either "under-ionized" or recombining plasma is constantly created. With sinusoidal modulation, there is a phase shift between the changes in current and voltage [5]. Thus, the description of processes in AP discharge is quite difficult.

Spectroscopic methods for studying plasma are one of the main methods for determining its parameters [2, 6]. This paper presents the results of studies of the dependence of the luminous efficacy of the First and Second Positive Systems of nitrogen lines (FPS and SPS) on the parameters of AP discharge. The spectral region for FPS is in the range $575-687nm$ and for SPS it is in the range $350-435nm$.

Selective amplification of individual spectral lines in the regions of the FPS and SPS bands of nitrogen, depending on the discharge parameters, is also reported. A frequency shift was detected in the emission of the strong red spectral line in pure nitrogen plasma and in the mixture of $CO_2:N_2:He=1:1:8$ in AP mode relative to the standard tabulated values at low pressures. The explanation of this phenomenon will be given in separate publication.

2. Experimental setup

When a glow discharge is fed with a modulated current containing constant and variable components, acoustic field appears in the discharge having the same frequency as the modulation. This acoustic field in the discharge tube interacts with the plasma, while the discharge tube behaves like an acoustic resonator. As a result of interaction with the acoustic field, the plasma goes into a new state - the AP one. In this new acoustoplasmic state, the plasma parameters can differ significantly from the plasma parameters without acoustic perturbations [4].

To work with a glow discharge, glass and quartz discharge tubes with cold tubular cathode and water cooling were manufactured. The diameter of the discharge channel is $5-10nm$ and the discharge length is $150-250nm$. The glass plates were glued at the ends of the tubes to receive visible radiation from the discharge along its axis. In order to receive the entire luminous flux, focons were attached to the ends of the tubes with a fiber connected to the spectrometer.

The diagram of the experiment is shown in Fig. 1.

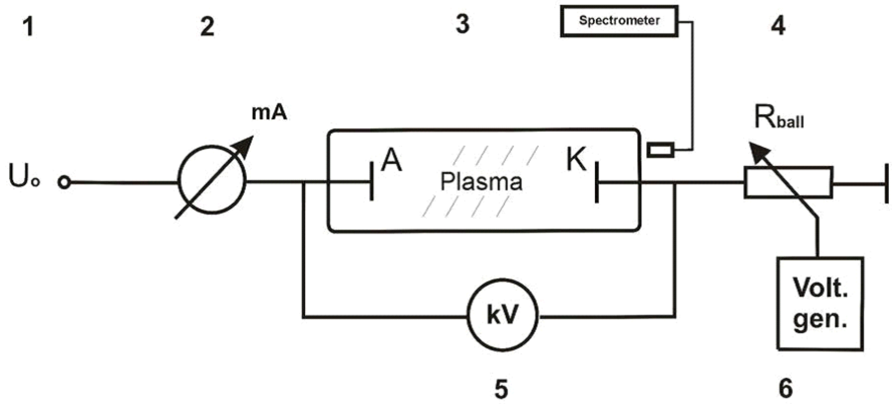


Fig. 1. The diagram of the experimental setup. 1 - source of constant high-voltage; 2 - milliammeter; 3 - discharge tube; 4 - spectrometer; 5 - controlled ballast resistance; 6 - kilovoltmeter; 7 - sound generator.

The source of constant high-voltage 1 is connected to the anode of the discharge tube 3. Using variable ballast resistance 5, which is connected to the cathode of the tube, the discharge current is modulated. As a variable ballast resistance, a high-voltage electron lamp was used. The lamp switching circuit, as a current generator, provided the possibility of obtaining a sinusoidally modulated current containing constant and variable components. The sound generator 7 provided the necessary frequency and amplitude of the current modulation. The magnitude of this current was measured with a milliammeter 2. The kilovoltmeter 6 showed the voltage applied to the gap anode-cathode of the discharge tube. The signals from milliammeter 2 and kilovoltmeter 6 were fed to digital indicators and a two-beam oscilloscope. Radiation from the cathode region of the discharge (perpendicular to the axis of the discharge) or from the end of the tube (from the side of the anode) was transmitted to the spectrographs 4 through the optical fiber. In this work, only the radiation from the anode side is considered. Different spectrographs were used to estimate the actual width and the position of the lines. The PC2000 OCEAN OPTICS company spectrograph, which had a resolution of $\pm 2nm$, was used as the main one. To increase the resolution of individual lines, we used computer spectrographs made in the laboratory on the basis of the ISP-51 prism spectrograph (resolution no worse than $0.4nm$) and on the basis of MDR (resolution better than $0.05nm$). In the visible region of the spectrum $(300-800nm)$, we studied the working mixture of a CO_2 laser $CO_2 : N_2 : He = 1:1:8$. The pressure of the gas mixture P_0 varied from 0.1 to $50 torr$. The constant component of the discharge current I_0 varied from 1 to $30mA$. The variable component of the discharge current $I\sim$ changes in the range from 0 to $30mA$. The modulation frequency of the discharge current varied from 0.1 to

$50kHz$. At $I \sim= 0mA$, there was a direct current discharge power mode (DC discharge); at $I \sim> 0mA$, there was an acoustoplasma discharge mode (AP discharge).

A multi-channel measuring complex was manufactured which allows using a standard multi-channel video surveillance and video recording system to simultaneously record electrical, optical and other plasma parameters, including the oscillograms [7].

During the experiment, due to the large flow of rapidly changing data, it is difficult to track the behavior of each of the discharge parameters. The manufactured measuring complex made it possible to conduct an experiment once, obtain a large database simultaneously for many parameters, and repeatedly reproduce the course of any part of the experiment by video recording. Thus, it was possible to investigate the relationship between the individual parameters of the experiment without repeating the actual experiment. Fig. 2 shows a diagram of the measuring complex.

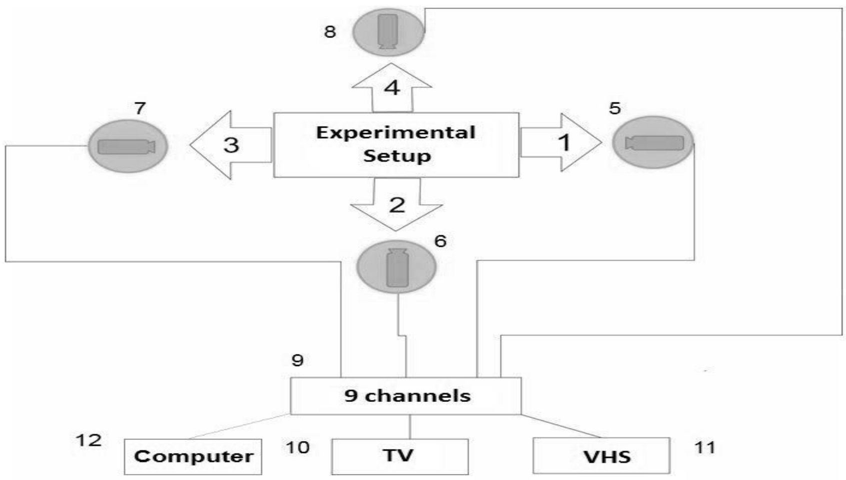


Fig. 2. The scheme of the measuring complex. 1-4 - information flows from the experimental setup; 5-8 - video cameras; 9 - nine-channel video surveillance system; 10 - TV monitor; 11 - video recorder; 12 - computer.

Arrows 1-4 represent the information flows during the experiment, which were simultaneously measured. Stream 1 - electrical parameters (discharge current, voltage across the anode-cathode gap, phase shift between current and voltage, etc.). Stream 2 - optical parameters (radiation intensity, intensity modulation, emission spectrum, etc.). Stream 3 - thermodynamic parameters (gas pressure in the tube, acoustic pressure, discharge temperature and tube walls). Stream 4 - geometric parameters (appearance of the discharge, its diameter, etc.). All data from digital and analog devices and all oscillograms were simultaneously recorded by 5-8 video cameras. Using the 9-channel video surveillance system 9, all channels were combined into one frame of the TV standard. These TV frames were received on a video monitor 10 and recorded in real time on a VCR 11 in the VHS standard and on the computer 12 in the MPEG-2 standard. From 4 to 8 hours of the experiment were recorded on one video cassette. To record to the computer, either several Web cameras or a TV tuner were used to input a complex video signal from a video surveillance system 9.

Figure 3 shows one of the experimental frames. The resolution of experimental situations in time was $40ms$ for neighboring frames and several μs inside each frame.

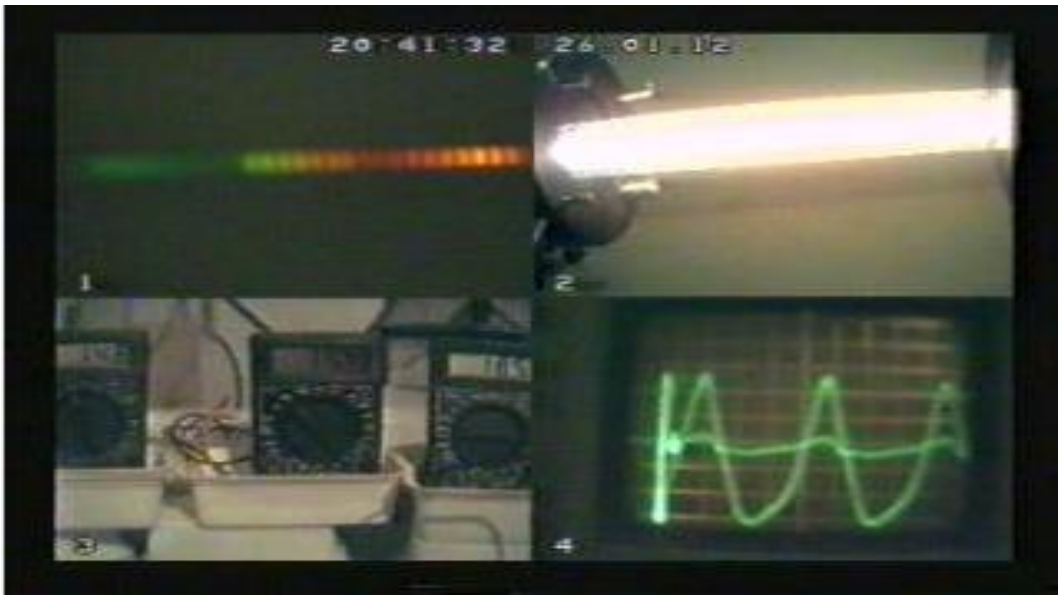


Fig. 3. One of the frames of the experiment on the monitor screen.

Figure 4 shows the emission spectrum of nitrogen AP obtained from the end of the discharge tube (along the discharge axis) using an OCEAN OPTICS PC2000 computer spectrograph. The abscissa shows wavelengths in nm. The ordinates show the intensities of the spectral lines (in arbitrary units). The gas pressure in the tube $P_0 = 10 torr$; constant component of the discharge current $I_0 = 8mA, I \sim= 8mA, f = 20\,kHz$; SPS - (second positive system) - the second positive nitrogen system; FNS - (first negative system) - the first negative system of the molecular ion of the nitrogen; FPS - (first positive system) - the first positive nitrogen system.

Typically, for light sources, luminous efficiency is measured in Lumens per Watt of power consumption (Lm/W), in this work the intensity of spectral lines is presented in arbitrary units (a.u.), and luminous efficiency K is presented in arbitrary units (Lm/W) (au) where W is the power applied to the gap anode-cathode.

In the AP mode, the current is modulated sinusoidally, and the power applied to the discharge depends nonlinearly on the current, therefore, some graphs directly depend on the discharge current.

3. Results and discussions

Our experiments showed that in the visible range of the spectrum of the $CO_2 : N_2 : He = 1:1:8$ mixture, the main role is played by the nitrogen spectrum, which accounts for $> 90\%$ of all the energy emitted in the visible spectrum. Therefore, in what follows we will talk about the lines of nitrogen only, remembering that we are dealing with a mixture.

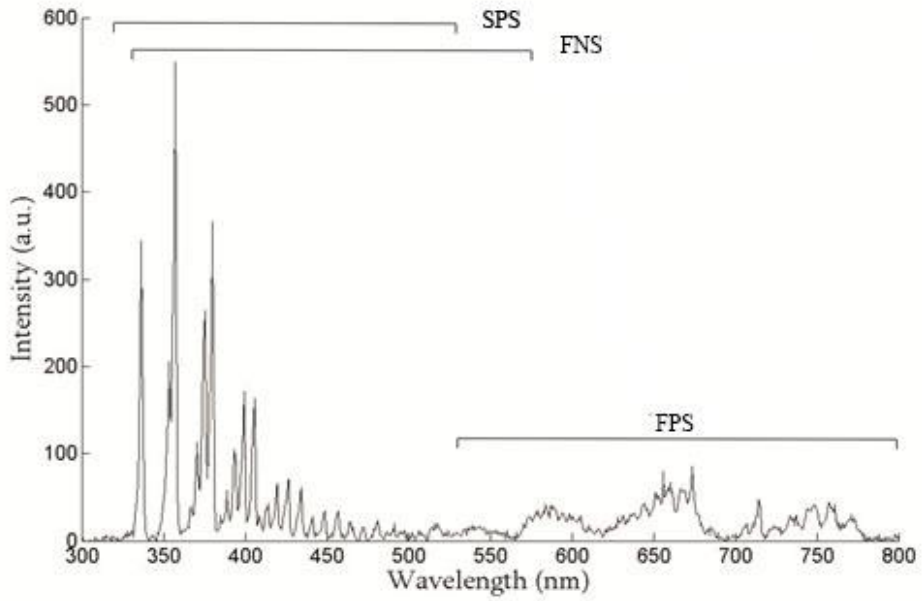


Fig. 4. The emission spectrum of nitrogen AP. P_0 = 10 torr, I_0 = 8mA; I~ = 8 mA; f = 20 kHz. The spectral lines included in SPS, FNS, FPS are indicated by brackets.

Figures 5-9 show the spectra obtained using the PC2000 OCEAN OPTICS for different pressures and frequencies of the discharge modulation. Fig. 5 corresponds to the DC mode, Fig. 6-9 – to the AP mode. It can be seen from the figures that in the AP mode, under certain discharge conditions, a single strong red line appears at about $655nm$ in the spectral region corresponding to FPS. The intensity of this red line decreases with increasing pressure and depends on the modulation frequency.

For example, for the pressure $4torr$, the intensity of the red line exceeds the intensities of the other FPS lines in the range of modulation frequencies of $0.1-0.2kHz$. With increasing modulation frequency up to $1\,kHz$, the intensity of the red line drops by 2 times. With the further increase in the frequency up to $7-8kHz$, the intensity of the strong line decreases even more. Then, in the region of $10\,kHz$, a sharp jump in the intensity of this strong red line occurs, it becomes several times larger than in the frequency range $0.1-8\,kHz$. With the further increase in the modulation frequency, the red line intensity decreases again. At the frequency $30\,kHz$ the red line intensity decreases 6 times, compared with the maximum. The red line has the highest intensity at the modulation frequency $10\,kHz$.

In the SPS region, emission lines with wavelengths $\lambda = 357.7\,nm$ and $\lambda = 380.5\,nm$ are the strongest ones. The strongest line is for wavelength $\lambda = 357.7\,nm$. The intensity of the SPS lines slowly decreases with increasing pressure and it is almost independent of the modulation frequency in the AP mode.

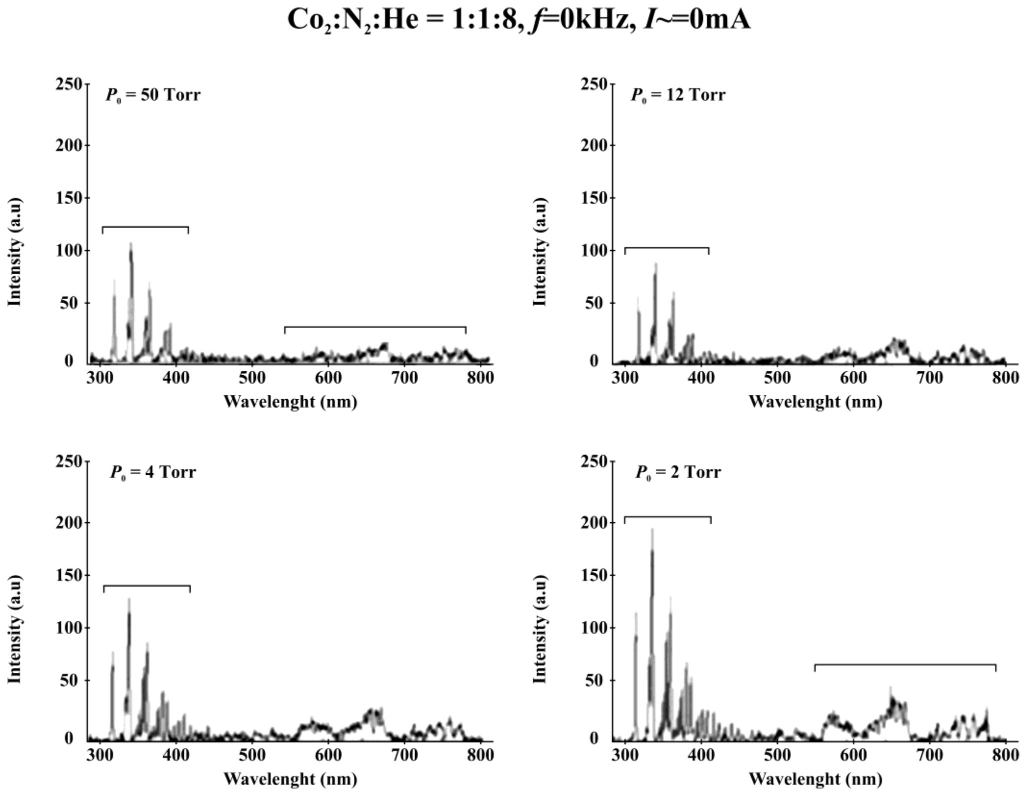


Fig. 5. Emission spectra of a mixture of CO_2: N_2: He = 1: 1: 8 in DC mode for different pressures. The brackets indicate the spectral bands of nitrogen SPS and FPS.

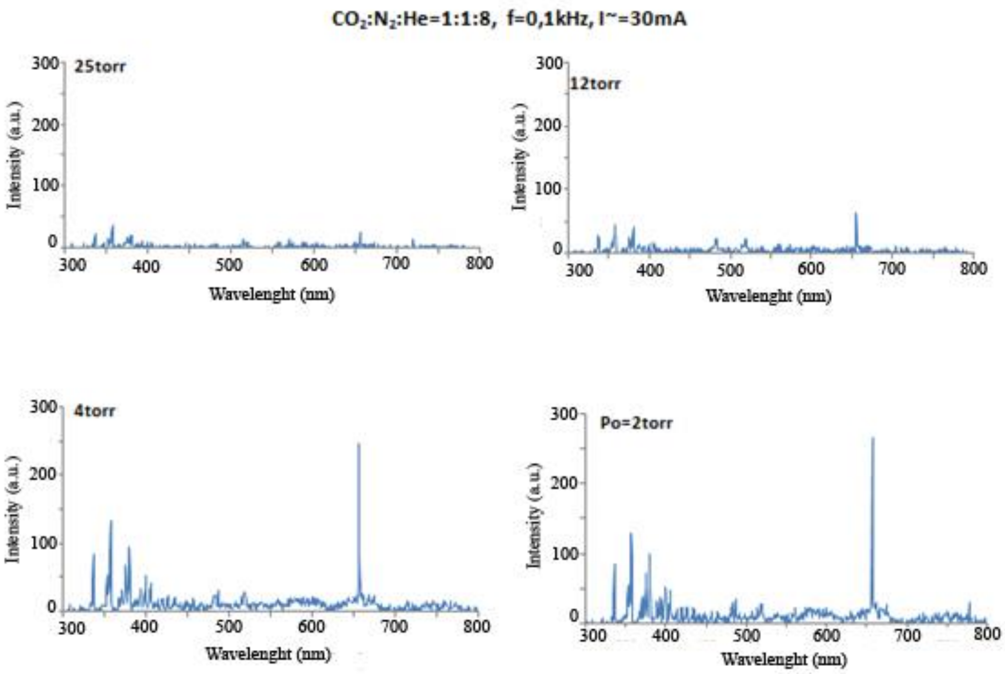


Fig. 6. Emission spectra of a mixture of CO_2: N_2: He = 1: 1: 8 in the AP mode for the frequency modulation current 0.1 kHz.

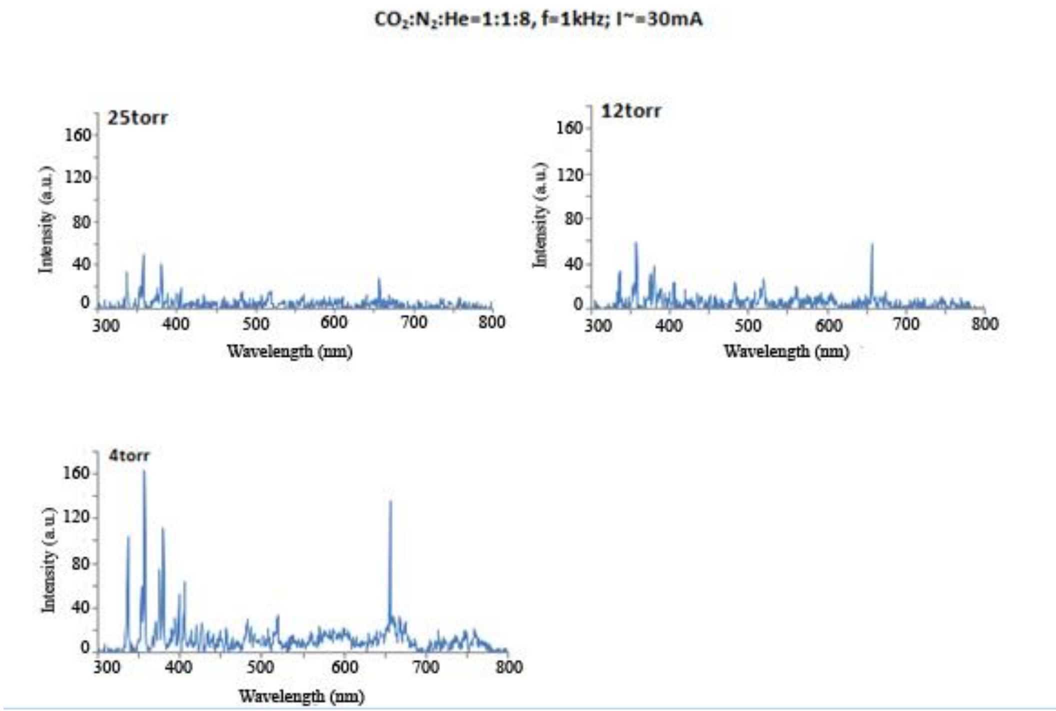


Fig. 7. Emission spectra of mixture of CO_2: N_2: He = 1: 1: 8 in the AP for frequency modulation current 1 kHz.

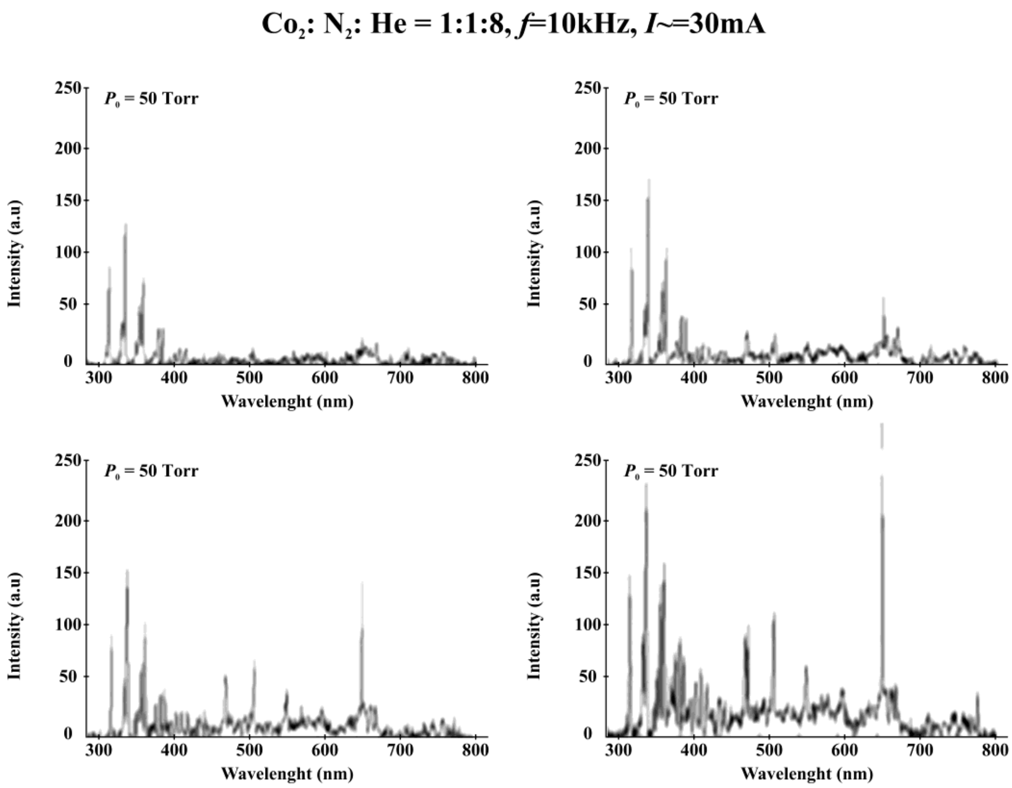


Fig. 8. Emission spectra of a mixture of CO_2: N_2: He = 1: 1: 8 in the AP for frequency modulation current 10 kHz.

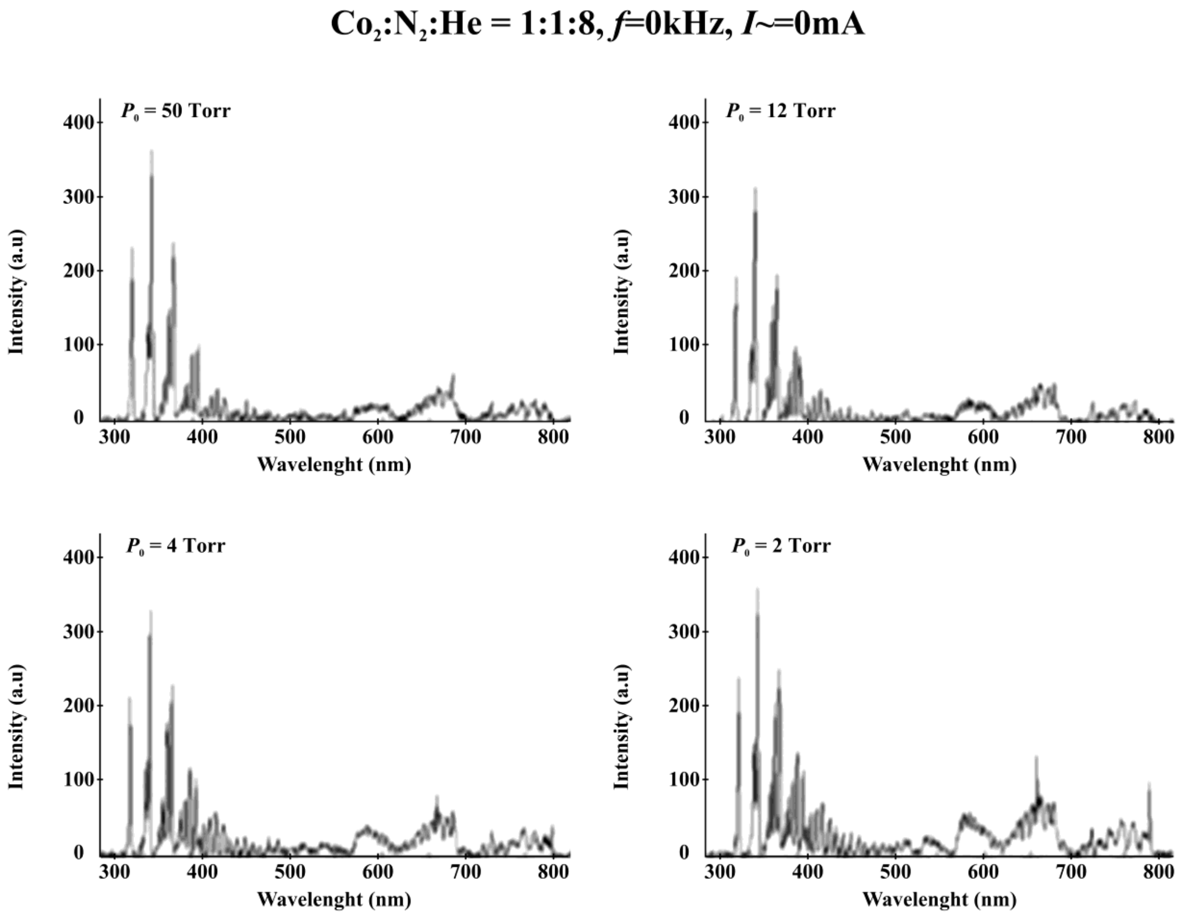


Fig. 9. Emission spectra of a mixture of CO_2: N_2: He = 1: 1: 8 in the AP mode for frequency modulation current 30 kHz.

Figure 10 shows the diagram of the levels of electronic and vibrational states, taken from [2].

Fig. 11 shows the part of fig. 10, where the regions of the appearance of strong lines and the bands of spectral lines of FPS ($\lambda = 575 - 687\ nm$, limited by thin green lines) and the region of SPS lines ($\lambda = 350 - 434.4\ nm$ are limited by thin purple lines) are marked. Thick red lines mark the area between the lines $\lambda = 662.4nm$ (1) and $\lambda = 654.5nm$ (2), the area in which a strong red line appears. Thick blue lines indicate the spectral lines $\lambda = 380.5nm$ (3) and $\lambda = 357.7nm$ (4).

Table 1, taken from [8] shows the main spectral lines observed in nitrogen discharge. v' and v'' are the upper and lower quantum vibrational levels.

Using tab. 1 and fig. 11, we can say that the strong red line is located between the vibrational quantum levels (7-4) and (6-3) for the corresponding electronic transitions $B^3\Pi_g$ and $A^3\Sigma_u^+$.

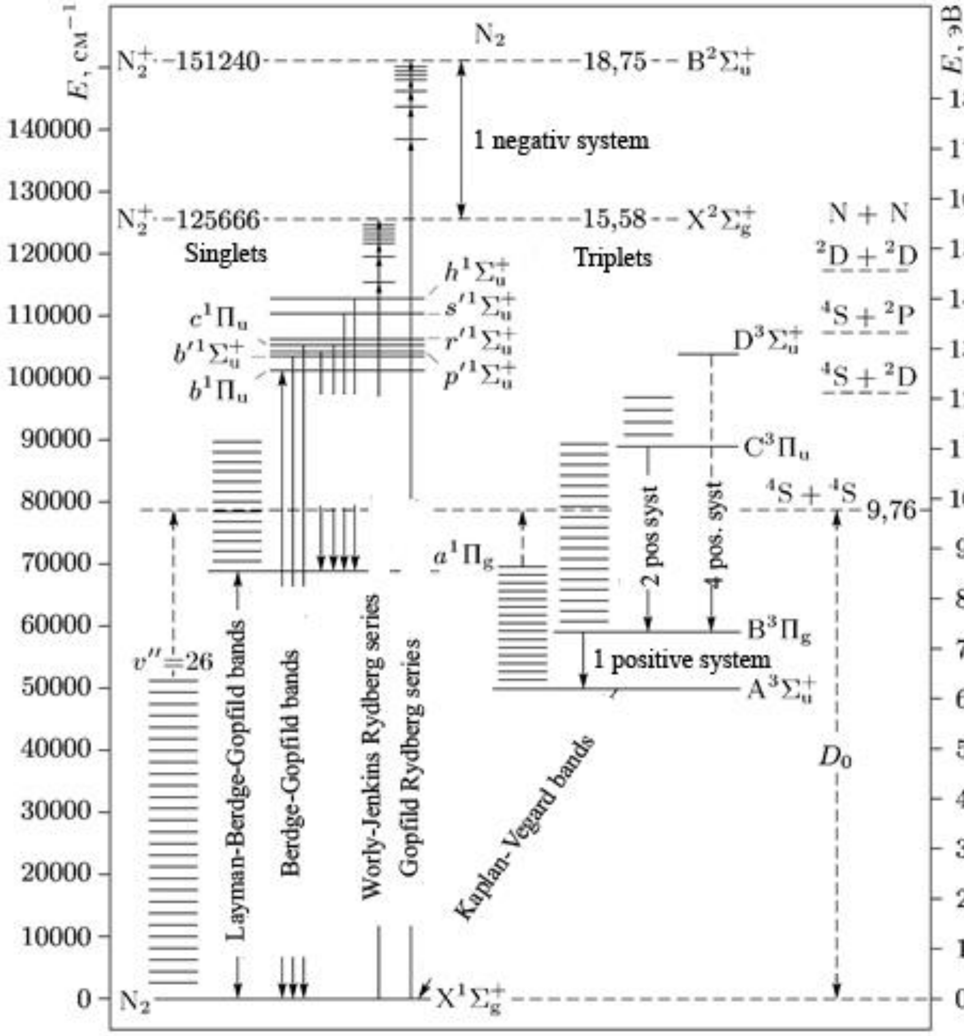


Fig. 10. Diagram of the levels of electronic and vibrational states for a nitrogen molecule [2].

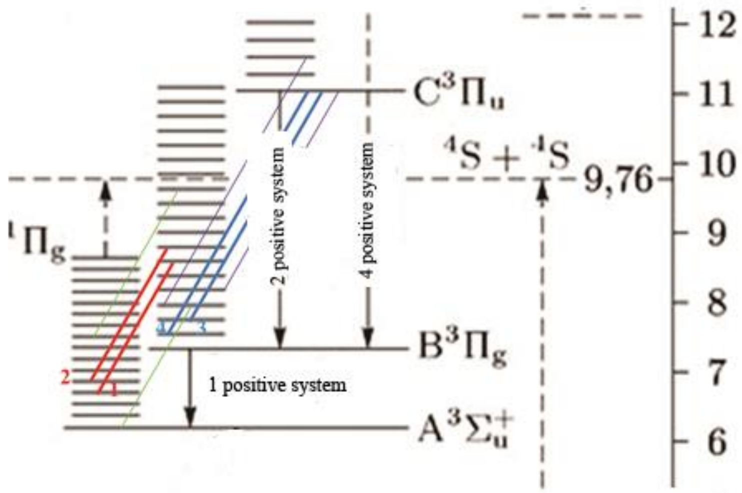


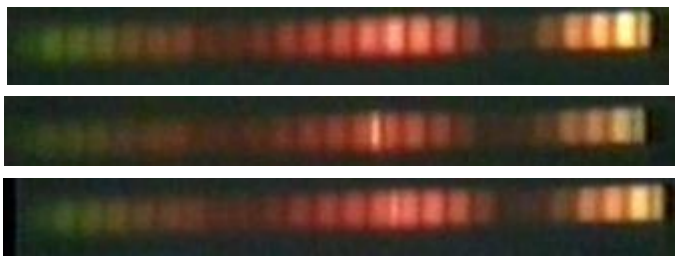
Fig. 11. 1 - λ = 662.4 nm; 2 - λ = 654.5 nm; λ = 575 - 687 nm
3 - λ = 380.5 nm; 4 - λ = 357.7 nm; λ = 350 - 434.4 nm.

Table 1. The main spectral lines observed in the discharge of molecular nitrogen [8]. SPS is the second positive nitrogen system; FPS is the first positive system. FNS is the first negative system (for an ionized nitrogen molecule N_2^+).

System	Energy of radiating states above the ground state (eV)	Sequence Δv	Band v'-v''	Wavelength
$N_2(C^3\Pi_u, v_C \rightarrow B^3\Pi_g, v_B)$	11.1	-1	2-3	350.05
			1-2	353.67
(SPS)			0-1	357.69
		-2	4-6	364.17
			3-5	367.19
			2-4	371.05
			1-3	375.54
			0-2	380.49
		-3	4-7	385.79
			3-6	389.46
			2-5	394.30
			1-4	399.84
			0-3	405.94
		-4	4-8	409.48
			3-7	414.18
			2-6	420.05
			1-5	426.97
			0-4	434.36
$N_2(B^3\Pi_g, v_B \rightarrow A^3\Sigma_u^+, v_A)$	7.4	4	12-8	575.52
			11-7	580.43
(FPS)			10-6	585.44
			9-5	590.60
			8-4	595.90
			7-3	601.36
			6-2	606.97
			5-1	612.74
		3	11-8	625.28
			10-7	632.29
			9-6	639.47
			8-5	646.85
			7-4	654.48
			6-3	662.36
			5-2	670.48
			4-1	678.86
			3-0	687.50
$N_2^+(B^2\Sigma_u^+, v_{Bi} \rightarrow X^2\Sigma_g^+, v_{Xi})$	18.7	0	2-2	385.79
			1-1	388.43
(FNS)			0-0	391.44
			2-3	419.91
		-1	1-2	423.65
			0-1	427.81

Figure 12 shows the spectra obtained on the converted ISP-51 spectrometer.

It is clearly seen from Fig. 12 that a strong red line appears between the standard spectral lines of nitrogen. Standard nitrogen lines look like rectangles because they represent vibrational-rotational bands for each vibrational level. It can be seen that the resulting strong red line is much more intense and much narrower.



a

b

c

Fig. 12. Spectra in the region of the FPS band for different modulation frequencies.
a) 1 кХз: b) 0.1 кХз: c) 10 кХз.

To determine the width of this narrow line, MDR diffraction monochromator was used, in which photomatrix was installed instead of photographic plate, i.e. it was also converted into the computer spectrograph with resolution of the order of $0.5\,\overset{o}{A}$. Figure 13 shows the spectrogram obtained on such a monochromator. It is clearly seen that a strong red line is located between the corresponding vibrational rotational bands. The color of the spectrogram is changed due to the color rendering of the receiving photomatrix.

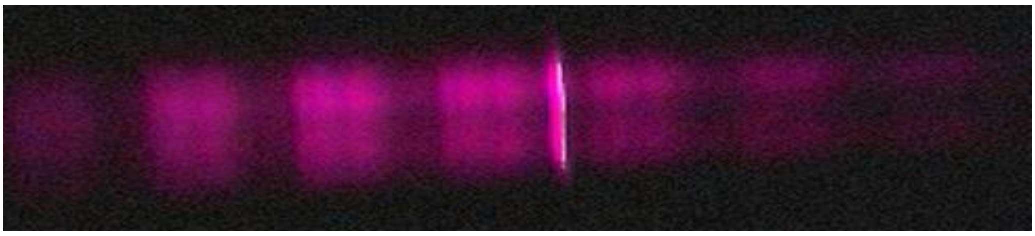


Fig. 13. CO_2: N_2: He = 1: 1: 8; P_0 = 2 torr; f = 0.1 kHz; I_0 = 21 mA; U = 2.58 kV; I~ = 17.2 mA.

Measurements on such a high-resolution spectrograph made it possible to determine the width of the strong red emission line, at half maximum it equals $0.3 \pm 0.1\,nm$ for the mixture pressure $0.1\,torr$.

Our measurements showed that for a DC discharge, the light efficiency $K\left(Lm/W\right)$ (a.u.), calculated for the entire FPS band, weakly depends on the discharge current I. It slowly grows with increasing current and can be represented by a linear dependence

$$K = aI + b \tag{1}$$

where I is the value of direct current. For the coefficients one has $a \ll b$. For example, for mixture of gases $CO_2 : N_2 : He = 1:1:8$ with pressure $P_0 = 10\,torr$, when the discharge current changes from 5 to $30mA$, the coefficient a is 100 times smaller than the coefficient b. Thus, all changes in K obtained in AP discharge can be attributed to AP effects and not to the change in the magnitude of the current during the modulation period.

In the AP mode, all the curves are well approximated by a power-law dependence

$$K = A(P_0)^{-B} \tag{2}$$

where A changes 2 times and B - 3 times, depending on the modulation frequency. Figure 14 shows the dependence of K on pressure P_0 and modulation frequency f for gas mixture of $CO_2 : N_2 : He = 1:1:8$ for the modulation depth $M = 1\,(M = I \sim / I_0$ is the ratio of the amplitude of the variable current component $I \sim$ to the constant current component $I_0)$.

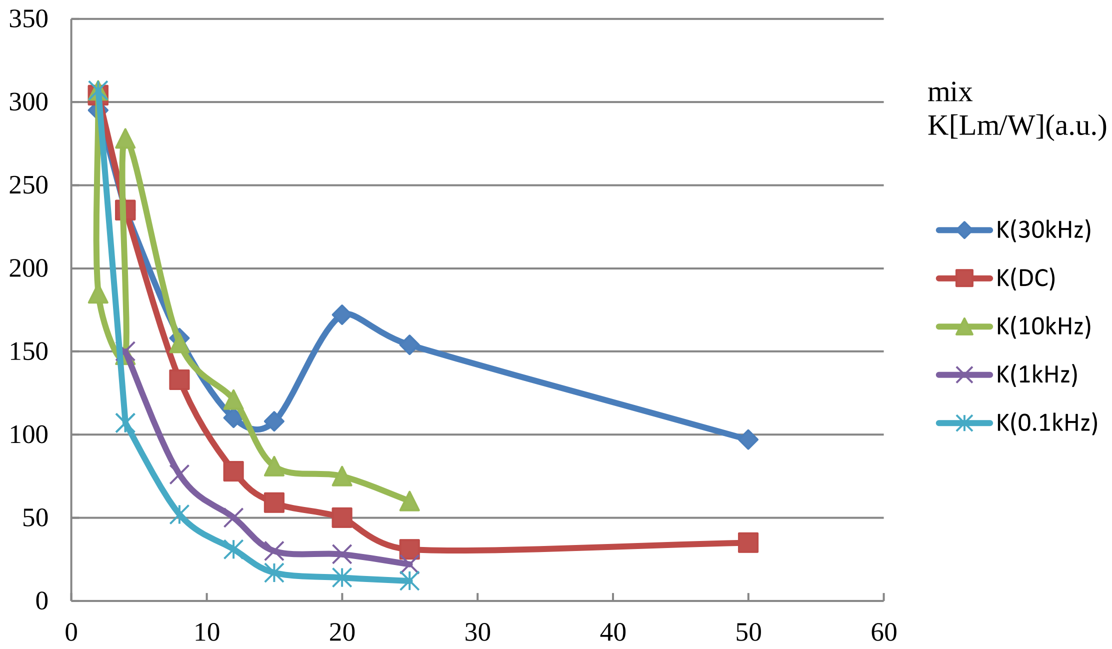


Fig. 14. Dependences of the light efficiency K on the pressure P_0 and the modulation frequency f at the modulation depth $M = 1$.

The hump in the curve corresponding to modulation frequency of $20-30\,kHz$ at pressures $>20\,torr$ is associated with the appearance of strong longitudinal AP striations.

The dependence of the light efficiency K on the modulation frequency is given, as a rule, by a polynomial of the $3rd$ degree. The minimum of the curve is in the frequency range $3-4\,kHz$ and the maximum is in the frequency range $10-20\,kHz$. The ratio of maximum to minimum equals 3-5.

For pure nitrogen, the above dependences are similar, only the absolute values change. As mentioned above, in the experiments sharp increase in the intensity of individual spectral lines of nitrogen was observed in the FPS and SPS bands, especially in the $CO_2:N_2:He$ mixture. Figure 15 shows the results only for the FPS band for the wavelength range of $600-700nm$ obtained on a PC2000 OCEAN OPTICS spectrometer.

Figure 15 clearly shows the resonance dependence of the intensity of the strong red line on the modulation frequency. The wavelength of the strong line is in the range $654-660nm$. When the discharge is supplied with direct current, the excess intensity of the strong red line is $1.3-1.5$ times greater than that for neighboring lines. At the modulation frequency $f=0.1\,kHz$ - 5-6 times larger; at $f=10\,kHz-17$ times larger; at $f=30\,kHz-1.7$ times larger.

Figure 16 shows the dependence of the intensity of the strong red line in the region of 655 *nm* on the pressure and modulation frequency. The pressure is limited to 20 *torr*, but in the experiment the pressure was changed to 300 *torr* and this increases the operating voltage on the discharge tube. Therefore, in this experiment, a discharge tube with discharge gap of 150*mm* in length and a discharge channel diameter of 10*mm* was used. For remaining experiments, discharge tubes with discharge gap 250*mm* were used. This explains the difference when comparing the curves of Figs. 14 and 16.

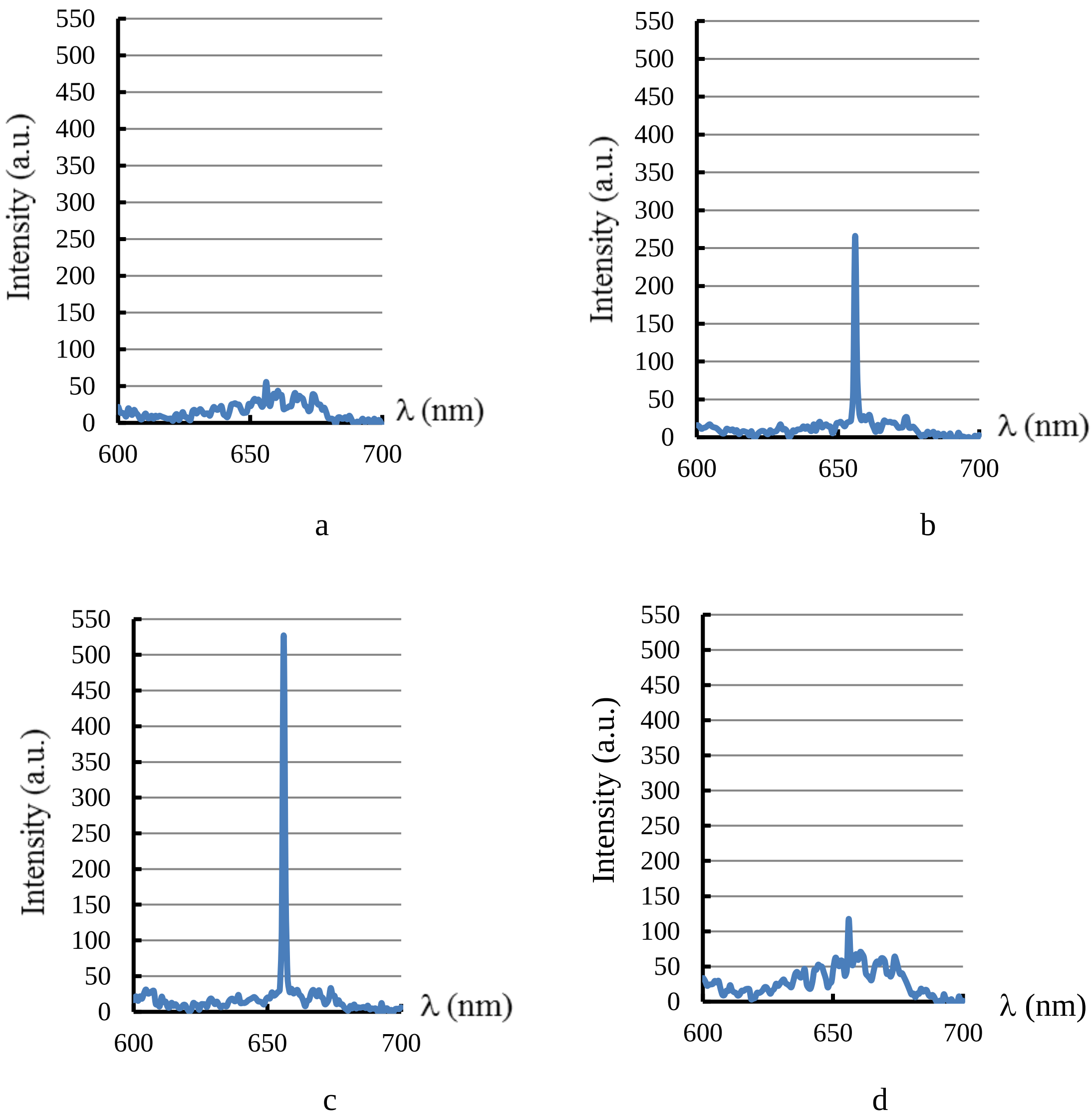


Fig. 15. The change in intensity (in arbitrary units) of the strong red line on the modulation frequency. The part of the FPS spectral region of nitrogen (600-700 nm) is presented in a mixture of CO_2: N_2: He = 1: 1: 8; pressure P_0 = 2 torr; I_0 = 20mA; *M* is 1; a) DC; b) *f* = 0.1kHz; c) *f* = 10 kHz; d) *f* = 30kHz.

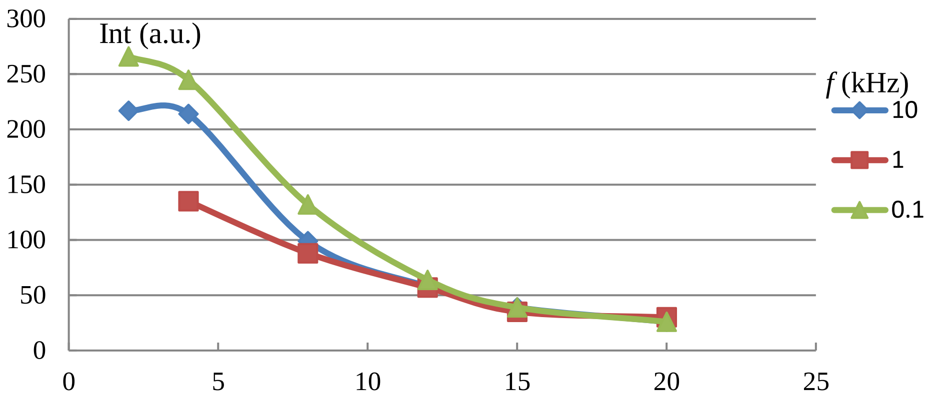


Fig.16. The dependence of the intensity of the strong red line for 655 nm on the pressure and modulation frequency.

It can be seen from Fig. 16 that the dependence of the intensity on the supply frequency, i.e. the resonance properties of AP practically manifest themselves only at low pressures $(<10\, torr)$.

Possible reasons for the appearance of this strong red line in the acoustoplasma mode will be the subject of a separate publication.

4. Conclusion

1. The AP mode of the discharge tube with a mixture of gases $CO_2 : N_2 : He = 1:1:8$ was experimentally studied. The emission spectrum of the discharge was studied in the visible region $(300-700nm)$. The dependences of the intensities of individual spectral lines and light efficiency in the bands of the First and Second Positive Systems of nitrogen lines (FPS and SPS) were studied. The corresponding spectral regions correspond to $574-687nm$ for FPS and $350-435nm$ for SPS.
2. It was found that, at certain values of the parameters, a strong narrow line appears in the discharge with the AP in the red region of the spectrum, in the region of $655nm$. The intensity of this strong red line can be 17 times higher than the intensity of neighboring lines of the emission spectrum. The width of this line at half maximum is $0.3 \pm 0.1nm$.
3. The intensity of the strong red line depends on the frequency and has resonant character. The maximum intensity is observed at a modulation frequency of the discharge current $10\, kHz$.
4. The strong red line appears between the standard spectral lines of the nitrogen FPS band.
5. Possible reasons for the appearance of this strong red line will be the subject of another publication.

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