MEASUREMENT OF ROTATIONAL TEMPERATURE OF MOLECULAR NITROGEN IN THE ANODE AREA OF NEGATIVE CORONA IN AIR UNDER TRICHEL PULSE MODE

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Experimental results of investigations of emission spectra of the second positive system of molecular nitrogen from the anode area of negative corona discharge in air are presented. In the Trichel pulse mode, the radiation intensity distribution in the electronic-vibrational-rotational $C^3\Pi_u(0)$-$B_3\Pi_g(0)$ transitions of molecular nitrogen is analyzed. Based on the analysis of the rotational structure of the spectral lines, taking into account the Boltzmann distribution of the rotational levels population, the rotational temperature (~ 470 K) of the molecular nitrogen in the anode area is determined. A theoretical calculation of emission spectra of the R-branch rotational lines is carried out.

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INTRODUCTION

In previous works [1, 2] it was shown that in the "needle-sphere" electrode system, along with the well-known glow near the needle cathode, there is also an anode glow. The radiation from the anode area (~ 1 mm from the surface of the spherical electrode) is observed. It has been experimentally established that the existence of radiation near the anode is also related to the processes that take place at the cathode. The authors suggested the existence of a perturbation wave in the region of increased field strength near the anode area, which was experimentally proved. The radiation intensity increases monotonically as the wave front moves deeper into the discharge gap toward the anode. At the same time, the intensity of radiation from the anode area is significantly low than the radiation intensity from the area of the needle cathode. The radiation from the anode area allows to realize various optical methods for diagnostics of a gas discharge characteristics. Spectroscopic studies of the radiation of a gas discharge make it possible to determine not only the local characteristics of the discharge (in particular, the electron distribution function), but also the temperature of the heavy gas particles. It is of interest to measure the rotational temperature of gas molecules, which value is equal to translational temperature. By determining the temperature from the anode area of the discharge, one can obtain information on the intensity of the ionization processes and analyze the effect of the gas temperature on the processes responsible for the transition of the discharge to the spark-breakdown stage. This will provide additional information for understanding the mechanisms of the formation of a stable sequence of the Trichel current pulses, and also for improving existing theoretical models of discharge in the nonstationary stage of burning.

1. EXPERIMENTAL SETUP

The object of the study in all experiments was a negative corona discharge in air at atmospheric pressure in the "needle-sphere" electrode geometry. The discharge was maintained in the Trichel pulse mode. The rotational temperature value was determined from the electron-vibrational-rotational transitions of the emission spectrum of the second positive nitrogen system. Investigations of the emission spectra from the anode area in the wavelength range 300…400 nm were carried out at the setup schematically shown in Fig. 1.

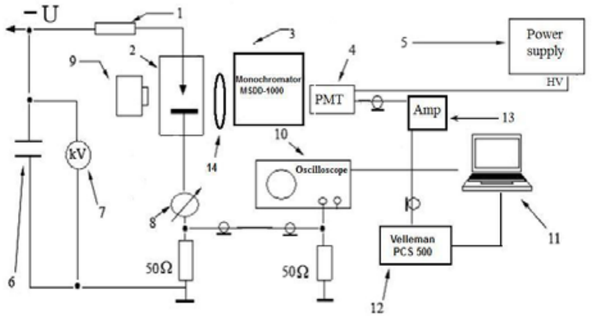


Fig. 1. 1 – ballast resistor R = 130 kΩ; 2 – discharge chamber; 3 – monochromator spectrograph SolarTii MSDD-1000; 4 – photomultiplier (PMT)-Hamamatsu R9110; 5 – stabilized high-voltage power supply for PMT; 6 – capacitive voltage filter of C = 1000 pF; 7 – HV voltmeter C196; 8 – micro ammeter M906; 9 – camera Olympus C 7070; 10 – oscilloscope Tektronix TDS-2024; 11 – computer PC; 12 – ADC Velleman PCS 500; 13 – DC amplifier IEC-CA3; 14 – fluorite (CaF_2) condenser

Stabilized adjustable high voltage power supply unit of 0…15 kV range was used to initiate the discharge burning. The voltage of the discharge gap was measured with a HV voltmeter. The average discharge current was measured with a micro ammeter. In the experiments, the electrode system "needle-sphere" was used. The cathode was a copper needle with a cross section diameter of 1 mm. The stainless steel anode was made in the form of a sphere with a diameter of 10 mm. The discharge was studied at the discharge gap of d = 7 mm. To measure the time and amplitude characteristics of the current pulses, calibrated current shunts with a nominal value of 50 Ω were used. The current shunts were calibrated using a Tektronix CT-1 calibration shunt with a signal bandwidth of 25 kHz to 1 GHz. A signal from the current shunts was analyzed with a digital oscilloscope 10 Tektronix TDS-2024B. The bandwidth of the oscilloscope was 200 MHz, sampling frequency 1 GS/s. Registration of radiation from the entire discharge gap was carried out with a digital camera.

Spectroscopic studies of the discharge were carried out at optical stand based on a double-dispersion monochromator-spectrograph "Solar-Tii" MSDD-1000. The registration of radiation from various areas of the discharge gap was carried out with a help of a slits system. To reach high spectral resolution, a double diffraction grating of 2.400 grooves / mm with a linear dispersion of 0.41 nm/mm was used. On the output slit of the monochromator, a high-speed photomultiplier Hamamatsu R9110 was installed. PMT characteristics: spectral sensitivity range of 185…900 nm, signal pulse rise time 2.2 ns. The signal from the PMT was transferred to the input of the DC amplifier IEC-CA3, which has the following characteristics: range of the conversion factor k – (10^{-10}…10^{-5}) A/B, the amplitude of the internal noise – not more than 1 pA, thermal drift of the output voltage – not more than 0.15 mV/deg. The signal from the amplifier was transferred to the input of the Velleman PCS 500 ADC, which was connected to a computer. The PC-Lab2000 software package allowed to display digitized data (visualize the spectrum) from the ADC Velleman PCS 500 on a computer monitor in real-time graphical mode, and also to record digitized data in the computer's memory.

2. EXPERIMENTAL RESULTS

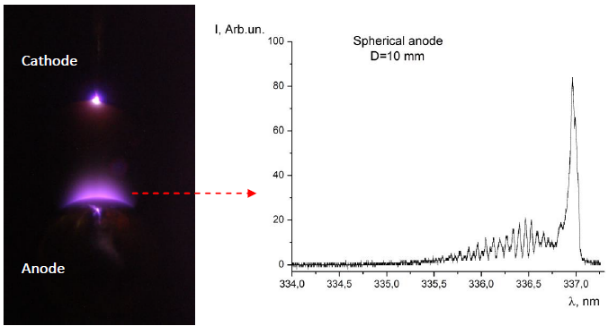
The spectra were recorded from the anode area of discharge zone (of ~ 1 mm from the spherical anode) during the burning of the discharge in the Trichel current pulse mode. In the wavelength range 300…400 nm, emission spectra corresponding to the second positive molecular-nitrogen system ($C^3\Pi_u$-B^3P_g transitions) were recorded [3]. The discharge gap was d = 7 mm. The monochromator slits were set to 0.05 mm (output) and 0.182 mm (input) with an inverse linear dispersion of the grating of 0.41 nm/mm.

2.1. DISCHARGE CURRENT CHARACTERISTICS

At experiments it is important to control the steady-state burning of the discharge. For this purpose, special treatment of the needle electrode was carried out before the experiments. Also the special electrode materials were selected and surface cleaning was carried out. The discharge current waveforms were registered in online mode with the oscilloscope. Also the current pulses repetition rate was measured. The steady-state repetition rate of the current pulses, and the unchanged shape of the current pulses waveforms, demonstrated the stable burning mode of the discharge.

2.2. OPTICAL AND SPECTRAL DIAGNOSTICS OF DISCHARGE RADIATION

Before carrying out the spectral measurements, the optimum diameter of the spherical anode, at which maximum radiation from the anode area is observed, was determined experimentally. It has been established that the optimal diameter of a spherical anode is 10 mm. Fig. 2 shows a photo of the discharge in the electrode system "needle-sphere" in the Trichel pulse mode (A), as well as a fragment (B) of the emission spectrum of the $C^3\Pi_u$ (0)-$B^3\Pi_g$ (0) transition in the wavelength range 334…337.3 nm. A spectrum was recorded from the anode area of the discharge gap. In the above photo, two clearly visible glowing regions are shown: cathode and anode.



a *b*

Fig. 2. Photo of the negative corona in the "needle-sphere" electrode system in the Trichel pulse mode. Exposure time 1 min (a); fragment of the emission spectrum of the $C^3\Pi_u(0)$-$B^3\Pi_g(0)$ transition (b). The average discharge current is I_c = 45 μA, the voltage applied to the discharge gap is U = 9.2 kV

To determine the values of the rotational temperature, the partially resolved rotational structure of $C^3\Pi_u$-$B^3\Pi_g$ transition spectrum was used. For the analysis of the spectra obtained in the experiment, it is necessary to use only separate lines of the R branch with range of rotational quantum numbers J = 20…29. In the remaining area of the spectrum of the electron-vibrational-rotational transition (0-0), the lines P, Q and R branches have superposition, which makes this region unsuitable for analysis.

For a more detailed analysis of the rotational structure of the spectrum, individual fragments of the spectrum were recorded at the maximum sensitivity of the ADC with respect to the input signal and with the use of an DC amplifier with a gain of up to k = 10^{10}. To increase the radiation intensity, due to a decrease in losses in the optical path, a short-focus fluorite (CaF_2) condenser was used. The registered fragment of the rotational structure of the spectrum in the wavelength range 334…335.6 nm is shown in Fig. 3. The spectrum is registered from the anode area of the negative corona in the Trichel pulse mode.

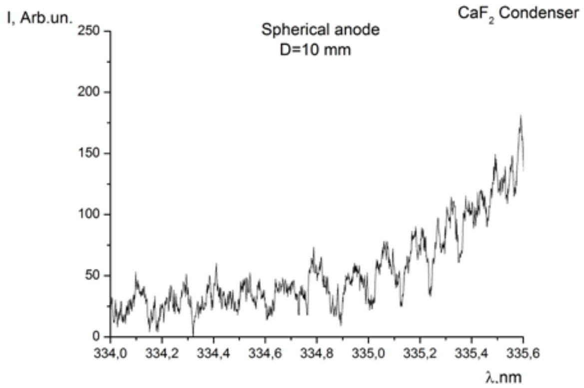


Fig. 3. Fragment of the emission spectrum in the wavelength range 334…335.6 nm. The average discharge current is I_c = 45 μA, the voltage applied to the discharge gap is U = 9.2 kV

Analysis of the obtained spectrum made it possible to determine the value of the rotational temperature of nitrogen molecules in the anode area of the discharge. To determine the correspondence of the wavelengths of the rotational lines to the rotational quantum numbers J, the Fortrat diagrams were calculated. For the electronic transition between $C^3\Pi_u$ and $B^3\Pi_g$ states, changes in the value ΔJ = -1, 0, +1, which generate P, Q and R spectrum branches, are allowed [4]:

$$\nu_{Q(J)} = \nu_0 + (B'_{v+1} - B''_v)J + (B'_{v+1} - B''_v)J^2, \tag{1}$$

$$\nu_{P(J)} = \nu_0 - (B'_{v+1} + B''_v)J + (B'_{v+1} - B''_v)J^2, \tag{2}$$

$$\nu_{R(J)} = \nu_0 + 2B'_{v+1} + (3B'_{v+1} - B''_v)J + (B'_{v+1} - B''_v)J^2, \tag{3}$$

where ν_0 is the wave number of the vibrational transition.

Calculation of the intensities of the rotational lines of electron-vibrational transitions was carried out taking into account the anharmonicity of vibrations of the nitrogen molecule and in the approximation of a non-rigid rotator for rotational energy

$$F_j = B \cdot J \cdot (J+1) - D \cdot \left[J \cdot (J+1)\right]^2 \tag{4}$$

The radiation intensity of an individual electron-vibrational-rotational band is determined from expression

$$I_{J'J''} = \frac{hc}{\lambda} \cdot N_{J'} \cdot A_{J'J''}, \tag{5}$$

where $N_{J'}$ – the population of the upper vibrational level, $A_{J'J''}$ – the transition probability, which are determined from expressions:

$$N_{J'} \sim \frac{B_e'}{kT_{rot}} \cdot (2J'+1) \cdot \exp\left(-\frac{B_e' J'(J'+1)}{kT_{rot}}\right), \tag{6}$$

$$A_{J'J''} = \frac{64\pi^4}{3h\lambda^3} \cdot \frac{S_{J'J''}}{2J'+1}, \tag{7}$$

where $S_{J'J''}$ is the intensity factor of Henle-London [4].

Thus, for the intensity of the rotational line of the electron-vibrational band, we obtain the following expression

$$I(\lambda) \sim \lambda^{-4} \cdot \frac{B_e'}{kT_{rot}} \cdot S_{J'J''} \cdot \exp\left(-\frac{B_e' J'(J'+1)}{kT_{rot}}\right). \tag{8}$$

It is important to note the change in the intensity ratio in the spectrum of P, Q, R branches, which must be taken into account when using experimental data to determine the rotational temperature of nitrogen molecules.

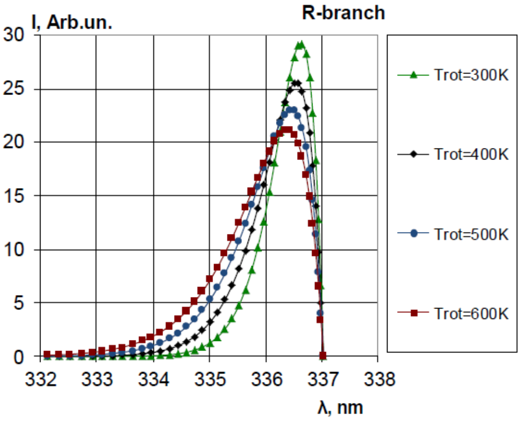


Fig. 4. Calculated spectrum of the distribution of the relative intensities of the R branch with a change in the rotational temperature in the range from 300 to 600 K. The transition is $C^3\Pi_u(0)$-$B^3\Pi_g(0)$

Below in Fig. 4 the calculated emission spectra of the rotational lines of the R branches of the electron-vibrational transition are shown. The spectra are calculated for different values of rotational temperature from 300 to 600 K in 100 K steps.

In the wavelength range 332…335 nm, the R-branch dominates in the spectrum, while in the region of the cant the lines thicken, and, therefore, the branches have superposition. It is important to note that R-branch spectrum have region without superposition, which is convenient for analysis when determining the rotational temperature by the relative intensity of the lines. In the Boltzmann distribution of the rotational levels population of excited electron-vibrational state, there is a simple relationship between the experimentally measured line intensity and the rotational temperature T_{rot} of the excited electron-vibrational state

$$\ln\frac{I(\lambda)}{\nu^4 S_{j'j''}} = -\frac{hc}{kT^*_r} \cdot F(j') + const, \tag{9}$$

where *F (j´)* is the energy of the upper rotational level in cm^{-1}, k is the Boltzmann constant, and *c* is the speed of light.

The linear dependence $\ln\frac{I(\lambda)}{\nu^4 S_{j'j''}}$ on *F (j´)* is an experimental confirmation of the existence of the Boltzmann distribution of the rotational levels population. It is important to note that the very weak intensity of the rotational lines of the R-branch significantly complicates analysis. Thus a large amplification of the output signal from a PMT is required. In this case, it is necessary to increase the signal-to-noise ratio and to gain a large statistical array of data for processing and analysis. Fig. 5. shows the Boltzmann plot obtained from linear approximation (using the program package ORIGIN Pro8.5) of experimental data in the analysis of the R-branch lines of rotational structure of emission spectrum. The diagram shows the linear dependence $\ln\frac{I(\lambda)}{\nu^4 S_{j'j''}}$ on F (J '). The slope angle of the straight line corresponds to the value of the rotational temperature of nitrogen molecules.

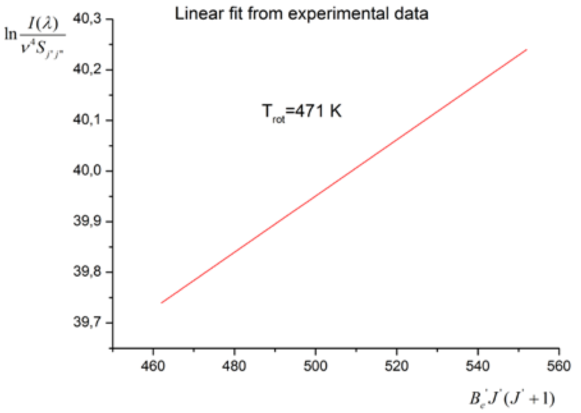


Fig. 5. Boltzmann plot obtained from the analysis of the spectra of the electron-vibrational transition of $C^3\Pi_u(0)$-$B^3\Pi_g(0)$ in the wavelength range 334…335.6 nm. The average discharge current is I_c = 45 μA, the voltage applied to the discharge gap is U = 9.2 kV

It is established from the plot shown in Fig. 5 that the rotational temperature of the nitrogen molecules in the anode area of the discharge is T_{rot} ~ 470 K. Such value of temperature indicates the area of low-temperature plasma in which efficient plasma-chemical processes can occur. At the same time, the burning mode of the discharge is high stable. To determine the conditions for the transition of the discharge to the stage of a spark breakdown, and also to optimize the electrode system, additional investigations are required. In addition, it is necessary to check the change of the gas temperature in the discharge gap over a wide range of voltages applied to the electrode system. Such methodological tasks require additional research.

CONCLUSIONS

In order to determine the principle of the stable sequence of Trichel current pulses, and to optimize the geometry of the electrode system, the emission spectra of the second positive nitrogen system from the anode area of the discharge are investigated. The distribution of radiation intensity in electron-vibrational-rotational bands corresponding to molecular nitrogen transitions from state $C^3\Pi_u(0)$ to state $B^3\Pi_g(0)$ is analyzed. A theoretical calculation of the intensity of rotational lines in the non-rigid rotator approximation is carried out. Based on the analysis of the rotational structure of the spectral lines, the rotational temperature (~470 K) of the molecular nitrogen in the anode area of discharge is determined.

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ИЗМЕРЕНИЕ ВРАЩАТЕЛЬНОЙ ТЕМПЕРАТУРЫ АЗОТА В ПРИАНОДНОЙ ОБЛАСТИ ОТРИЦАТЕЛЬНОЙ КОРОНЫ В ВОЗДУХЕ В РЕЖИМЕ ИМПУЛЬСОВ ТРИЧЕЛА

О.В. Болотов, В.И. Голота, Ю.В. Ситникова

Приведены результаты экспериментальных исследований спектров излучения второй положительной системы молекулярного азота из прианодной области отрицательной короны в воздухе в режиме импульсов Тричела. Проанализировано распределение интенсивности излучения в электронно-колебательно-вращательных переходах $C^3\Pi_u(0)$-$B_3\Pi_g(0)$ молекулярного азота. На основе анализа вращательной структуры спектральных линий, с учетом больцмановского распределения заселенности вращательных уровней, определена вращательная температура молекул азота (~470 K) в прианодной области разряда. Проведен теоретический расчет спектров излучения вращательных линий R-ветви вращательной структуры спектра.

ВИМІРЮВАННЯ ОБЕРТАЛЬНОЇ ТЕМПЕРАТУРИ АЗОТУ В ПРИАНОДНІЙ ОБЛАСТІ НЕГАТИВНОЇ КОРОНИ В ПОВІТРІ В РЕЖИМІ ІМПУЛЬСІВ ТРИЧЕЛА

О.В. Болотов, В.І. Голота, Ю.В. Сітнікова

Наведено результати експериментальних досліджень спектрів випромінювання другої позитивної системи молекулярного азоту з прианодної області негативної корони в повітрі в режимі імпульсів Тричела. Проаналізовано розподіл інтенсивності випромінювання в електронно-коливально-обертальних переходах $C^3\Pi_u(0)$-$B_3\Pi_g(0)$ молекулярного азоту. На основі аналізу обертальної структури спектральних ліній, з урахуванням больцманівського розподілу заселеності обертальних рівнів, визначена обертальна температура молекул азоту (~470 K) у прианодній області розряду. Проведено теоретичний розрахунок спектрів випромінювання обертальних ліній R-гілки обертальної структури спектра.