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VUV spectra of diffuse plasma formed by colliding streamers

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Abstract. In this report we carry out studies of the VUV emission of diffuse discharges in different gases formed in gaps with non-uniform electric field by sub-nanosecond voltage pulses due to run-away electrons and colliding streamers. VUV emissions in mixtures of pure rare gases and its mixtures with hydrogen and nitrogen in measured in a gap between two needles. The data on Ar_2* and Xe_2* emission both in diffuse and contracted discharges are consistent with the results of our previous work and fundamentally differ from the spectral measurements described in some reports. It was found that VUV emission intensity of H_2 Lyman band in the range of 149–161 nm increases significantly in mixtures with neon.

Gas mixtures were identified in which powerful emission is observed on the lines of atomic nitrogen at 149 and 174 nm. An assumption was made about the possibility of obtaining stimulated radiation on these lines.

Keywords: VUV emission, non-uniform electric field, diffuse plasma, colliding streamers.

1. Introduction

In recent years, interest has increased in the study of collisions of leaders and streamers during pulsed discharges in air and other gases of various pressures. Streamers were studied in the tip-to-tip [1] and blade-to-blade intervals [2]. The collision of the leaders was observed during the propagation of negative lightning towards the Earth [3], as well as in the air at atmospheric pressure at meter intervals [4]. In [5, 6], the interaction of diffuse jets in long dielectric tubes filled with low-pressure gases was studied.

In recent works [7, 8], it was shown that the run-away electrons initiates the development of wide streamers (ionization waves) and the diffuse discharges are formed after the streamer closed the gap. Therewith the diffuse discharge does not have time to transform into spark when the short voltage pulses are used. It should also be noted a large number of computational works simulating physical processes in gas discharges in an inhomogeneous electric field, in which important regularities in the formation of ionization waves have been established.

The purpose of this work is to study the collision of plasma diffuse jets and streamers, shape of plasma formations in the interaction region and the plasma VUV emission spectra.

2. Experimental setup and measurement techniques

An experimental setup for studies of the spectral and amplitude-time characteristics of the diffuse discharge is shown in Fig. 1. A discharge gap formed by two needles or one needle and plane 0.4 or 6 cm long, placed in a chamber (2), was connected to high-voltage pulsed generator (3) with adjustable amplitude via a 3 meter long 75 Ω cable (4). The chamber was attached to a VM-502 vacuum monochromator (Acton Research Corp.) (6) equipped with an EMI9781B (PMT) photomultiplier tube (7) made it possible to record both the spectral distribution of the discharge plasma radiation energy in the region of 100–545 nm and its temporal characteristics. The minimum rise time of the radiation pulse reliably recorded by the PMT is ~ 3 ns, and the fall time is 30 ns. The monochromator was pumped out with an NMDO-0.1 (NORD-100) oil-free pump up to a residual pressure of 10^{-6} Torr.

A GIN-55-01 generator produced voltage pulses of negative polarity with the amplitude in the incident wave of ~ 38.5 kV with the duration of $\tau_{0.5} \approx 0.7$ ns (FWHM) and a rise time of $\tau_{0.1\text{-}0.9} \approx 0.7$ ns. For the case of using the GIN-55-01 generator. The high-voltage generators were triggered by a BNC565 master generator (5). Voltage pulses from the generators were applied to the discharge gap via a 3-m-length cable (4). This made it possible to record radiation pulses from the diffuse discharge within 30 ns after applying the voltage pulse.

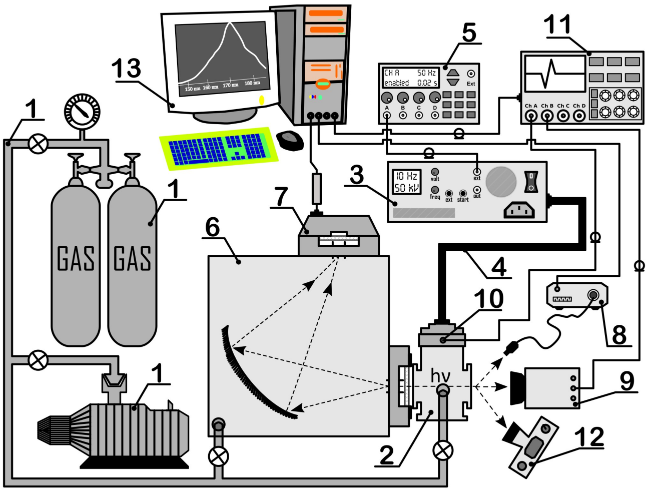

Fig. 1. Schematic diagram of the experimental setup: 1 – gas pumping system; 2 – discharge chamber; 3 – high-voltage pulsed generator; 4 – high-voltage cable; 5 – triggering generator; 6 – vacuum monochromator VM-502; 7 – PMT EMI9781B; 8 – spectrometer StellarNet EPP2000C-25; 9 – photodiode FEK-22; 10 – capacitive voltage divider; 11 – digital oscilloscope; 12 – digital camera; 13–PC.

Besides, spectra and waveforms of the radiation pulses in the visible and UV ranges were recorded with an EPP2000C-25 spectrometer (StellarNet Inc.) with a known spectral sensitivity (8) and a FEK-22 vacuum coaxial photodiode with a temporal resolution of ~ 1 ns (9), respectively. Voltage pulses were registered with a capacitive voltage divider (10). Waveforms of the electrical signals were recorded with digital oscilloscopes Tektronix TDS-3054B (500 MHz, 5 GS s^{-1}) (11).

3. Experimental results

The main part of research was carried out with maximal voltage pulse amplitude from the generator, operating with a pulse repetition rate of 10 Hz. The diffuse discharge was formed in this case, the gap breakdown occurred within a few tenths of ns, and the discharge plasma resistance rapidly decreases to a small value (tenths of Ohm). Radiation parameters of diffuse discharge in Ar are shown in Figs. 2–5.

Time-integrated images of diffuse discharge plasma emission in air, nitrogen and helium at atmospheric pressure taken per one implementation are shown in Fig. 2. The diameter of the brightly glowing region of the wide streamer is commensurate with the gap size. A darker region is visible in the middle of the gap. It is caused by a decrease in the excitation rate of nitrogen molecules when an increase in the streamer diameter. When the streamer reaches its maximal diameter, the electric field strength at its front is minimal. After bridging the gap, the reverse streamer propagates from the grounded electrode toward the tip one and produces excited nitrogen molecules whose emission is observed in the figures. The short duration of the voltage pulse limits the length of the reverse streamer. Nevertheless, bright anode spots appear due to an increase in the discharge current density in the vicinity of the anode. The size of bright anode spots is affected by the kind of gas used. Note that the appearance of bright spots on the anode can be influenced by an increase in the electric field between the front of the first streamer and the flat anode.

Emission spectra of diffuse discharge in rare gases and hydrogen are shown in Fig. 3. The second VUV continuum of R_2* dimers, where R=Ar, Kr, Xe, with peaks at 126, 146 and 172 nm, respectively, dominates in the range 100–520 nm. Continuum emission at 220–270 nm is found to evident in all rare gases. Mainly this radiation comes from the near-electrode regions, where the current density is very high and the glow of streamers developing from the needles is visible. The intensity of broadband radiation in the wavelength region above 350 nm increased with increasing rare Xe pressure. This, in turn, can be explained by a decrease in the duration of the diffuse phase of the discharge. Small additions of Xe in Ar lead to almost complete disappearance of the Ar_2^* radiation radiation at 126 nm and the appearance of bands of ArXe* with peak at 146 nm and Xe^*_2 dimers at 172 nm.

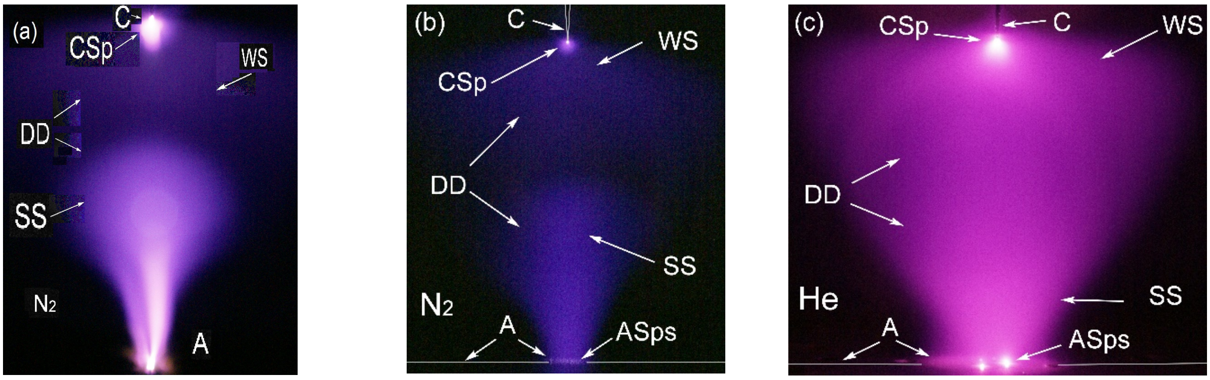

Fig. 2. Time-integrated images of the discharge plasma emission in a 6 cm pin-to-pin (a) and pin-to-plane (b-c) gaps filled with (a-b) nitrogen and (c) helium at atmospheric pressure. C – cathode (pin), A – anode, CSp – cathode sport, ASps – anode sports, WS – wide streamer, SS – second streamer, DD – diffuse discharge.

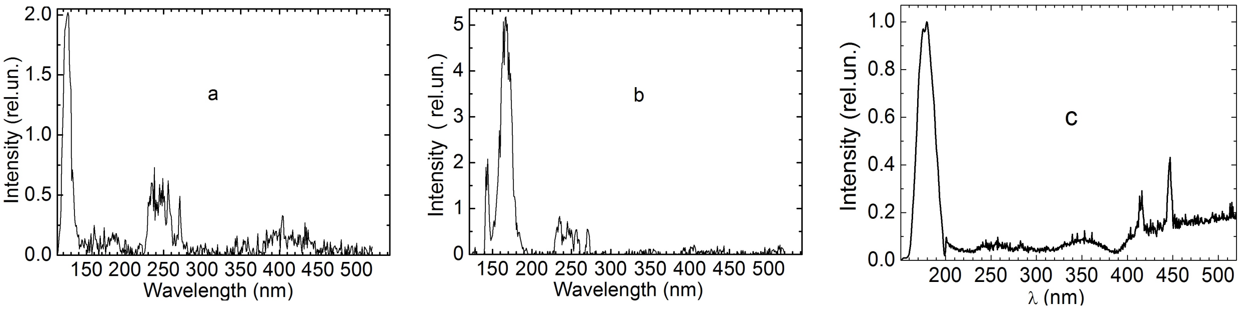

Fig. 3. VUV spectra of the diffuse plasma between two needles in Ar at 2 atm (a), Ar:Xe=2.25 atm:1.5 Torr gas mixture (b) and Xe at 3 atm (c).

Spontaneous emission of the diffuse discharges in pure hydrogen and its mixtures with some rare gases are shown in Fig. 4. The discharge has the form of weakly luminous in the visible spectral range diffuse channels starting from the needles and than quickly expanding towards the middle of the gap, forming a glow without signs of arc channels. Bright arc channels appear against the background of diffuse glow in gas mixtures with argon. As a result, intensity of UV and visible emission in mixture with Ar becomes more noticeable. The intensity of the UV and visible glow in the discharge balk slightly increased with He additions, while the intensity of visible radiation becomes more noticeable in mixtures with neon. Additions of helium and argon reduced the intensity of the VUV emission of the discharge plasma, as well.

It is seen from Fig. 4 that the Lyman band of H_2 in the wavelength range 120–170 nm is most intense in the VUV spectra of the diffuse discharge plasma in pure H_2 and its mixtures with Ne. Intensities of the atomic hydrogen lines at 100–120 nm were low compared to that of the Lyman band. The Lyman band intensity increases with decreasing hydrogen pressure, and its maximum is observed at a pressure of about 100 Torr. This is due to an increase of the overvoltage across the gap during the diffuse discharge formation by nanosecond high-voltages pulses when H_2 pressure is decreased. Consequently, the *E*/*p* parameter across the discharge gap increases which improves population rate of $B^1\Sigma^+_u$ level of hydrogen molecule. Increase in H_2 pressure results in the fast fall-off of the average electron energy and strongly decreases the overall efficiency of $B^1\Sigma^+_u$ level population.

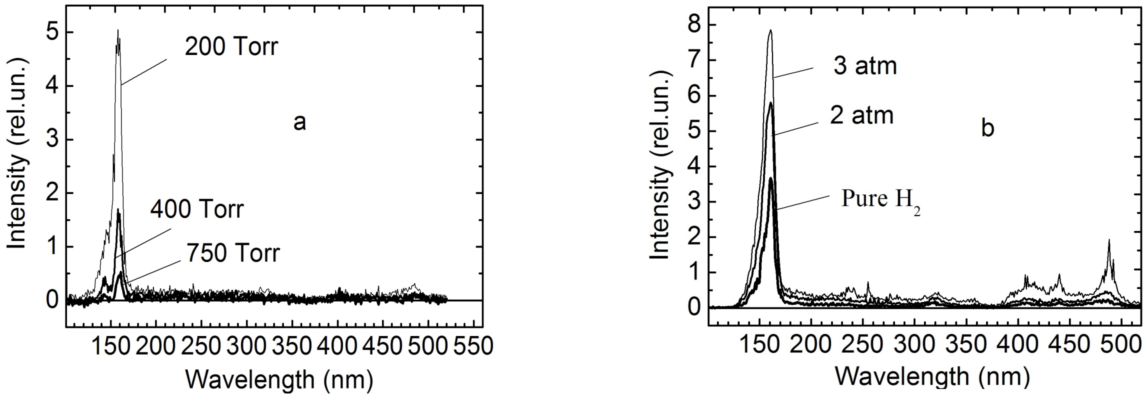

Fig. 4. Spectra of the diffuse discharge in pure H_2 at different pressures (a) and in mixtures of 100 Torr H_2 with Ne addition (b).

It was found that neon sufficiently improved the intensity of Lyman band. Since the intensity of spontaneous emission *P*(*t*) is proportional to the population of the upper level *n* of $B^1\Sigma^+_u - X^1\Sigma_g^+$ transition and the probability of spontaneous emission *A*:

$$P(t) = n\,[B^1\Sigma^+_u]\,A, \tag{1}$$

It follows from Fig. 4 and (1), that Ne increase population of $B^1\Sigma^+_u$ level hydrogen molecule.

The buffer gas effect can be explained in several ways. A volume discharge in neon-hydrogen mixtures had been widely used to pump a laser on Ne transition at $\lambda = 585.3$ nm. It is believed that in the kinetics of this laser, hydrogen is used to clean the lower laser level in the Penning ionization reaction:

$$Ne^* + H_2 \rightarrow H_2^+ + Ne + e. \tag{2}$$

However, another reaction channel (2) can be considered. Significant fraction of the input electric energy is spent on the formation of excited Ne* atoms and Ne^*_2 excimer molecules in a volume discharges in Ne–H_2 gas mixtures at high Ne pressure. Then, in collisions with excited Ne atoms and molecules H^*_2 in higher lying states can be produced and population of the $B^1\Sigma_u^+$ hydrogen state is possible by cascades from the excited H^*_2 states.

Ne_2^* excimer emission on the second continuum which corresponds to transitions from $^3\Sigma_u$ excimer state to the repulsive ground state at wavelength range 76–88 nm with peak at 84 nm and first excimer continua due to the radiative decay of vibrationally excited levels of the $^1\Sigma_u$ excimer state, centered between 73 and 75 nm along with Ne* resonance lines at 73.59 and 74.37 nm are dominated in discharge plasma spectra in mixtures with neon [9]. Under the conditions of our experiment, the radiation of neon molecules and atoms will be effectively absorbed by H_2 molecules in the ground state forming highly excited H_2^*, and the process described above can again be repeated.

The $B^1\Sigma_u+$ level can also be populated in the processes of recombination of H_2^+ molecular ions and collisions of H_2 molecules with excited H* atoms, which are produced in the reaction:

$$Ne^*_2 + H_2 \rightarrow 2Ne + H + H^*. \tag{3}$$

To determine the physical reasons for the effect of Ne effect, additional calculations and studies are required. Note that we previously obtained a significant increase in the parameters of the H_2 laser operation on the Lyman band in mixtures with neon [10].

An interesting result was obtained when studying a diffuse discharge in mixtures of light inert gases with nitrogen (see Fig. 5). In the spectrum of the discharge in a number of mixtures of nitrogen with helium, neon and argon, very intense emission of atomic lines of nitrogen was detected. The intensity of these lines significantly exceeded the intensity of radiation on the second positive nitrogen system, which is not typical for diffuse discharge in mixtures with N_2 [11]. The VUV emission pulses repeated the discharge current pulses, which were observed in the gap with a period of 30 ns due to reflections of the initial voltage pulse from the gap and the generator. The intensity of the lines at 149 and 174 nm increases sharply with the voltage pulse amplitude. In [12] reported the possibility of obtaining amplification on the atomic nitrogen lines. To check the possibility of obtaining lasing at 149.5 and 174.5 nm, measurements of the VUV radiation of an extended diffuse discharge are planned, using experimental device with long blade electrodes [9].

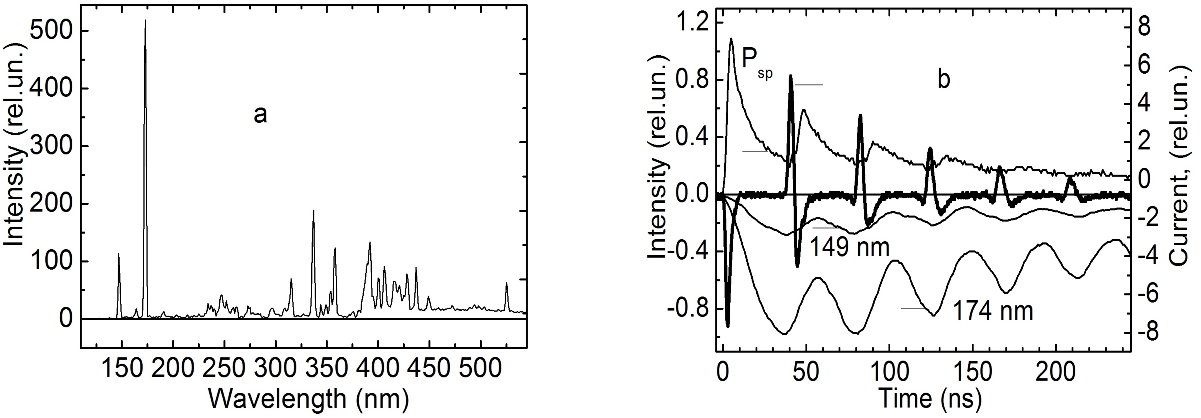

Fig. 5. Emission spectrum of a diffuse discharge in a He–N_2 gas mixture (a) and waveforms of pulses of visible and VUV radiation at 149 and 174 nm and discharge current (b).

4. Conclusion

Studies of emission in the VUV spectral region in Ar, Xe, H_2 and N_2 and its mixtures of a diffuse discharge formed by colliding streamers has been performed. In argon, the highest intensity is observed for the emission of Ar_2^* dimers with a maximum at a wavelength of 126 nm, even at a voltage pulse duration of 0.7 ns at half maximum. If small amount of xenon is added into argon, the Ar_2^* radiation almost completely disappears and the bands of $ArXe^*$ and Xe_2^* dimers appear. Xe_2^* dimers emission dominates in discharge in pure xenon, as well.

Continuum emission the UV range is found to evident in all rare gases. Mainly this radiation comes from the near-electrode regions, where the current density is very high and the glow of streamers developing from the needles is visible.

In hydrogen, emission on Lyman band with a maximum at 160 nm is observed in the diffuse discharge spectra and the luminescence intensity in the region of 220–280 nm is relatively low. If argon or helium is added into hydrogen, the Lyman band decreases. It was found that neon addition to H_2 significantly increases the VUV radiation intensity. Several physical mechanisms of the effect of neon additions have been proposed.

Very intense lines of atomic nitrogen at 149 and 174 nm were detected in the spectrum of the discharge in a number of mixtures of nitrogen with helium, neon and argon, which intensity significantly exceeded the intensity of radiation on the second positive nitrogen system. This is not typical for diffuse discharge in mixtures with N_2. The possibility of development of VUV laser operating on the atomic nitrogen lines is suggested.

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5. References

[1] W. Shi, Q. Zhang, Research on the breakdown process of needle-shaped electrode switch in air, *IEEE Transactions on Plasma Science*, vol. **46**, 358, Feb. 2018; doi: 10.1109/tps.2018.2790399

[2] A.N. Panchenko, D.A. Sorokin, V.F. Tarasenko, Gas lasers pumped by runaway electrons preionized diffuse discharge, *Progress in Quantum Electronics*, vol. **76**, 100314, March 2021; doi: 10.1016/j.pquantelec.2020.100314221

[3] E.M. Bazelyan, Yu.P. Raiser, *Physics of lightning and lightning protection.* Moscow: Fizmatlit, 2001. [in Russian].

[4] P.O. Kochkin, A.P.J. van Deursen, U. Ebert, Experimental study on hard x-rays emitted from metre-scale negative discharges in air, *Journal of Physics D: Applied Physics*, vol. **48**, 025205, Dec. 2015; doi: 10.1088/0022-3727/48/2/025205

[5] A.I. Shishpanov, D.O. Ivanov, S.A. Kalinin, Collision of ionization waves in long discharge tubes, *Plasma Research Express*, vol. **1**, 025004, May 2019; doi: 10.1088/2516-1067/ab1b8d

[6] V.F. Tarasenko, E.K. Baksht, V.A. Panarin, N.P. Vinogradov, Streamers initiated by a capacitive discharge at air pressure 0.2–6 Torr, *Plasma Physics Reports*, vol. **49**, 786, Jul. 2023; doi: 10.1134/S1063780X23700393

[7] V.F. Tarasenko, G.V. Naidis, D.V. Beloplotov, I.D. Kostyrya, N.Y. Babaeva, Formation of Wide streamers during a subnanosecond discharge in atmospheric-pressure air, *Plasma Physics Reports*, vol. **44**, 746, Aug. 2018; doi: 10.1134/S1063780X1808008

[8] V.F. Tarasenko, Runaway electrons in diffuse gas discharge, *Plasma Sources Sci. Technol.*, vol. **29**, 034001, Feb. 2020; doi: 10.1088/1361-6595/ab5c57

[9] N. Masoud, K.Martus, K. Becker, Vacuum ultraviolet emissions from a cylindrical dielectric barrier discharge in neon and neon-hydrogen mixtures, *Int. J. Mass Spectrometry*, vol. **233**, 395, April 2004; doi: 10.1016/j.ijms.2004.02.007

[10] A.N. Panchenko, V.F. Tarasenko, and V.V. Kozhevnikov, VUV lasers pumped by diffuse discharges, *Applied Physics B*, vol. **129**, 178, Oct. 2023; doi: 10.1007/s00340-023-08125-5

[11] Fierro, G. Laity, and A. Neuber, Optical emission spectroscopy study in the VUV-VIS regimes of a developing low-temperature plasma in nitrogen gas, *Journal of Physics D*, vol. **45,** 495202, Nov. 2012; doi: 10.1088/00223727/45/49/495202

[12] G.N. Gerasimov. E. Krylov, D.I. Stasel'ko, and I.V. Alekseev, Amplifying the VUV radiation of atomic nitrogen in helium, argon, krypton, and xenon, *Journal Optical Technology*, vol. **79**, 462, Aug. 2012; doi:10.1364/jot.79.000462 10.1364/jot.79.000462