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UV and VUV radiation of rare gases and nitrogen in diffuse discharges, formed in an inhomogeneous electric field

V.V. Kozhevnikov[1,2,*], *A.N. Panchenko*[1], *V.F. Tarasenko*[1,2]

[1]*Institute of High Current Electronics SB RAS, Tomsk, Russia*
[2]*National Research Tomsk State University, Tomsk, Russia*
[*]*vlad.kogevnikov@mail.ru*

Abstract. In this report we carry out additional studies of the VUV and UV emission of diffuse discharges formed by sub-nanosecond voltage pulses in rare gases and nitrogen. As a result, the optimal conditions for lasing on the second positive nitrogen system were determined and the data were obtained on of Ar_2* and Xe_2* emission both in diffuse and contracted discharges.
The data obtained are consistent with the results of our previous work and fundamentally differ from the spectral measurements described in some reports. Besides, parameters of the spontaneous and stimulated emission on the nitrogen second positive system and the conditions in which the emission is observed in is very different from the conditions under which this radiation has been usually produced.
Keywords: VUV emission, non-uniform electric field, diffuse discharge.

1. Introduction

A large number of works are devoted to investigations of discharges in inert gases and hydrogen, as well as in their mixtures. Exciting hydrogen and argon with a pulsed discharge and an electron beam create sources of laser and spontaneous radiation in the VUV spectral region. RF discharges in mixtures of hydrogen with noble gases are used in various industrial applications: etching processes, surface passivation, film deposition, photoresist stripping in the new low-k material strip technology and Extreme Ultra-Violet lithography for multilayer mirror cleaning. The interest in studying ion-molecular reactions in Ar/H_2 mixtures is also due to the use of these mixtures in fusion devices and the fundamental problem of excitation mechanism for the observed ArH^+ emission lines. Plasma chemistry in Ar/H_2 and $Ar/H_2/CH_4$ mixtures has also been studied in mW plasma-activated chemical vapor deposition reactors for diamond film deposition and a high-pressure dc arc jet.

Up to recent years, considerable attention has begun to be paid to pulsed discharges. Various gases have been studied out using gaps with sharply non-uniform electric field [1–3]. It was found that when voltage pulses of ns and sub-ns duration are applied to the gap of the tip-plane or tip-tip geometry, a diffuse plasma in pressured gases is formed due to preionization by run-away electrons formed in amplified electric field near electrodes with a small radius of curvature and bremsstrahlung X-rays [4]. In recent works [3, 4], 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.

2. Experimental setup

The emission and current-voltage characteristics of the diffuse discharge in the chamber shown schematically in Fig.1 are investigated. The chamber had two CaF_2 windows, in front of which there was a vacuum monochromator, a spectrometer, and an FEK-22SPU photocell or a camera.

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The best way to form a diffuse discharge with small interelectrode gaps with a maximum delay until sparking is to use both electrodes with a small radius of curvature. It was established earlier that the polarity of the voltage pulses did not significantly affect the formation of a diffuse discharge, and the negative polarity of an electrode of small curvature made it possible to easily detect a beam of runaway electrons.

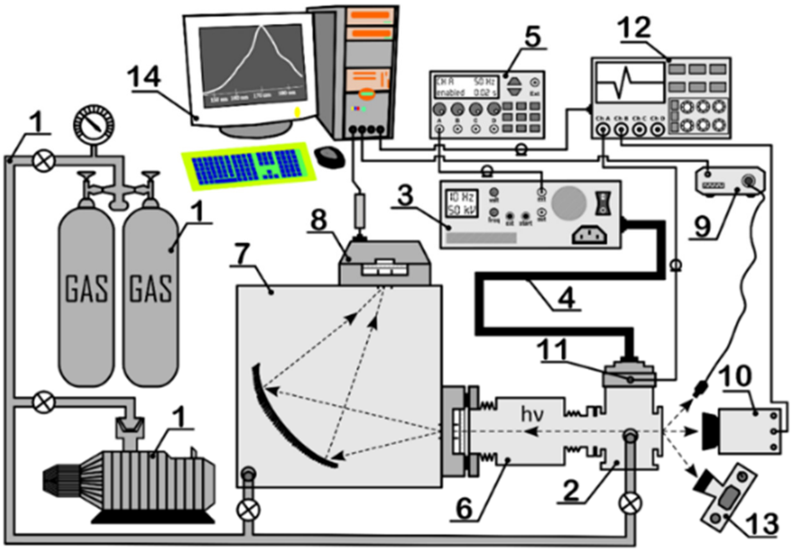


Fig.1. Schematic of the experimental set-up: 1 – gas pumping system; 2 – discharge chamber; 3 – high-voltage pulsed generator; 4 – high-voltage cable; 5 – triggering generator; 6 – vacuum monochromator VM-502; 7 – PMT EMI9781B; 8 – spectrometer StellarNet EPP2000C-25; 9 – photodiode FEK-22; 10 – capacitive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

Therefore, both electrodes we used were made from 5.6 mm segments. sewing needles with a base diameter of 0.75 mm and a radius of curvature of the tip of 40 μm. One electrode was attached to a cone, turning into a cylinder 6 mm in diameter, and the second electrode was attached to a flat grounded flange. Using two needle electrodes with a small distance between them can reduce the excited volume for both diffuse and narrowed discharges and, accordingly, increase the specific energy input.

The discharge glow was photographed with a digital camera. The emission spectra were recorded in the range from 200 to 800 nm using an EPP2000C-25 spectrometer (StellarNet-Inc.) with a known spectral sensitivity; In the range 120–540 nm, a VM-502 vacuum monochromator was used (Acton Researcher Corp.).

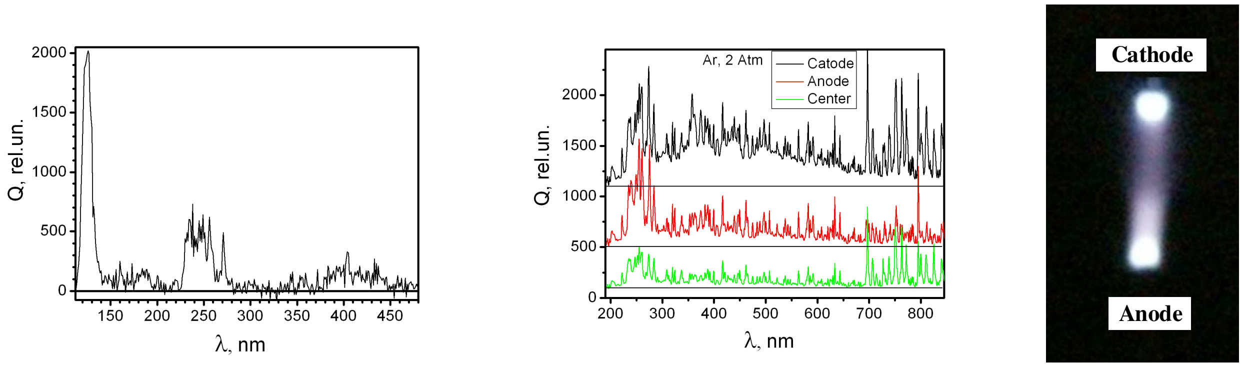
The temporal characteristics of radiation in some spectral ranges were measured using an EMI 9781 B photomultiplier tube, which allowed resolving the leading edge of a signal with a duration of ~3 ns and a trailing edge with a duration of ~30 ns, using an FEK-22SPU photodiode with a temporal resolution of ~1 ns.

3. Argon, hydrogen and its mixtures

3.1. Pure Ar

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 argon is shown in Fig.2.

As can be seen from Fig.2a, the radiation of the second continuum of Ar_2^* dimer has the maximum at the wavelength of 126 nm and dominates in the range 110–520 nm.



(a) (b) (c)

Fig.2. Emission spectra and image of diffuse discharge in argon at a pressure of 2 atm; (a) whole discharge region; (b) center and near-electrode regions of discharge gap; (c) plasma image.

This agrees with the results obtained in [5] and does not support the conclusions of [6]. The argon emission spectra from different discharge regions and integral photograph of the discharge gap and are presented in Fig.2bc. At a pressure of 2 atm one can see the region of diffuse discharge about 1 mm in diameter in the gap center and brighter radiation of the spark channel, whose intensity is higher near the needles, as shown in Fig.2c. The bright channel diameter was about 1 mm under these conditions.

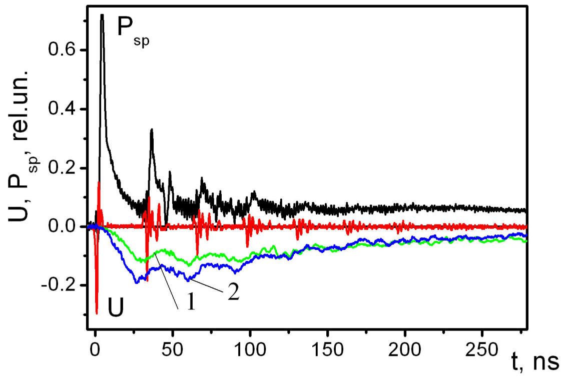


Fig.3. Waveforms of voltage (U) and time dependences of the discharge radiation pulse in the range 200–650 nm measured through the CaF_2 window (P_{sp}) and the intensity of radiation at 126 nm from a diffuse discharge plasma in pure Ar at 2 Atm (1) and in Ar:He=2:1 Atm mixture (2).

Continuum emission at 220–270 nm (see Fig.2a and Fig.2b) is found to evident in all gas pressured 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 anode and cathode is visible. The continuum includes lines of the electrode material, as well [7]. Diffuse luminescence is retained in the center of the gap; accordingly, the intensity of this continuum decreases. In the red region, there are strong lines of atomic argon formed in the dissociative recombination flow $Ar_2^+ + e^- \rightarrow Ar^{**} + Ar$; $Ar^{**} \rightarrow Ar^* + h\nu$. The broad-band UV-VIS emission can be is associated with free-free transitions $Ar^+/Ar + e \rightarrow Ar^+/Ar + e + h\nu$ (Bremsstrahlung emission) and free-bound transitions: $Ar^+ + e \rightarrow Ar^* + h\nu$, $Ar_2^+ + e \rightarrow Ar_2^* + h\nu$ (radiative recombination) [7].

Based on the two tip geometry experimental system, waveforms of voltage (U) and time dependences of the discharge radiation in the range 200–650 nm measured through the CaF_2 window (P_{sp}) and the intensity of radiation at a wavelength of 126 nm from a diffuse discharge plasma in Ar (1) and Ar-He (2) mixture are shown are shown and in Fig.3.

As can be seen in Fig.3, the Ar_2^* emission power begins to grow after the arrival of reflected pulses, which can be explained by the additional power input from them. The power of discharge radiation in the visible and UV spectral regions behaves similarly. Helium addition to Ar improves the Ar_2* emission power.

3.2. Ar-H_2 mixture discharge

Based on the tip-tips geometry, integral photographs of the diffuse discharge in H_2 at different pressure are shown in Fig.4a, while Fig.4b depicts spectra of the diffuse discharge plasma in pure hydrogen and its mixture with argon. Bright regions near the electrodes in pure H_2 are less noticeable compared to the discharge in argon. The glow region narrows with increasing hydrogen pressure. Accordingly, the luminescence intensity in the region of 220–280 nm is relatively low as shown in Fig.4b.

Similarly to [8] Lymam bands $B^1\Sigma^+_u \rightarrow X^1\Sigma^+_g$ with a maximum at ≈160 nm are dominated in he diffuse discharge spectra, and the emission intensity is maximal at hydrogen pressure of 150 Torr. The addition of argon leads to some increase in the intensity due to an increase in the breakdown voltage and, accordingly, the pumping power.

(a)

(b)

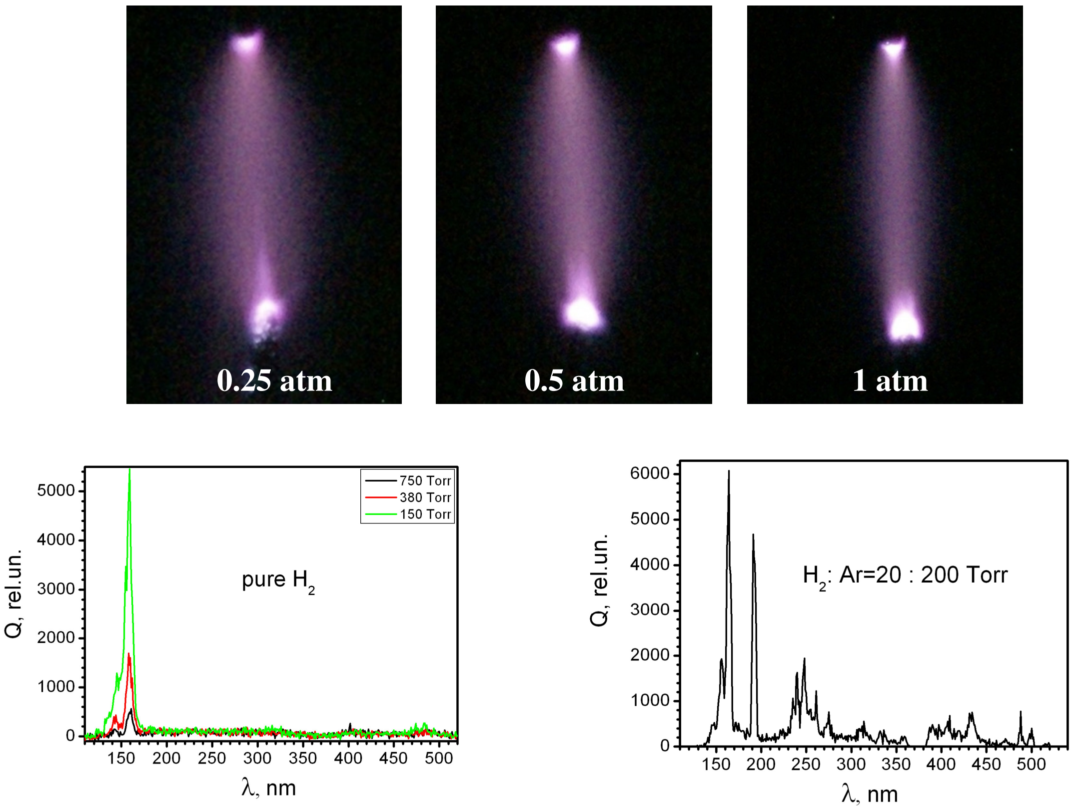


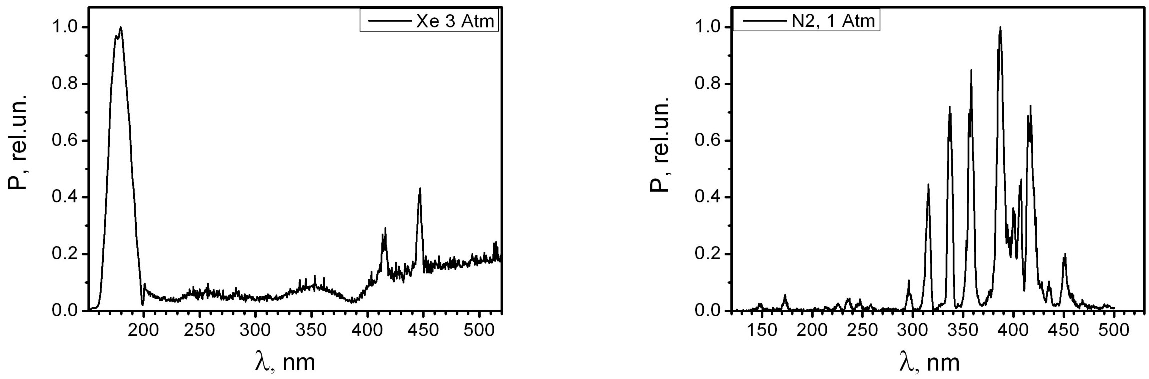
Fig.4. Plasma image and emission spectra of the diffuse discharge. (a) Integral photographs of the diffuse discharge in H_2 at different pressure (The cathode is located at the bottom). (b) Emission spectra of diffuse discharge. (Left: pure H_2; Right: H_2:Ar = 20:200 Torr gas mixture).

Argon additions to H_2 (H_2:Ar = 200:20 Torr) violate the uniformity of the diffuse discharge, which can be seen from the appearance of the continuum at 220–280 nm in Fig.4b. There is also a noticeable change in the spectrum. The luminescence band with peak at 160 nm narrows, the peak at 156 nm becomes more pronounced and strong line of Ar^+ ion at 191 nm appears in the discharge spectra.

The duration of the glow of the hydrogen bands is approximately 150–200 ns, which is much shorter than the duration of the glow of the argon dimer (see Fig.3).

4. Xe and nitrogen

The results obtained are shown in Fig.5. It is seen that in a pulsed diffuse discharge, the second continuum of rare gas dimers Xe_2* makes the largest contribution to the VUV radiation energy. In the case of N_2 only week VUV lines are evident, while emission on C-B band of N_2 and that of B-X band of CN are dominated in the UV and visible ranges. Powerful lasing at 337 nm was obtained only with 30 cm long blade electrodes. Efficient VUV lasing on F^*_2 and H_2 molecules was obtained, as well, in mixtures of F_2 and H_2 with rare gases.



(a) (b)

Fig.5. Spectra of diffuse discharges in Xe at 3 Atm (a) and N_2 at 1 Atm (b).

Thus, the studies performed confirm that the main part of the energy emitted by the plasma of a diffuse xenon discharge in the range of 120–850 nm is provided by the spectral transitions of xenon dimers.

Broadband radiation in the visible and ultraviolet spectral regions from spark discharge is mainly associated with recombination radiation, which is widely used in high pressure xenon lamps.

The above data are in good agreement with our previous results obtained for a voltage-initiated discharge of pulses with an amplitude of 220 kV and at half maximum (in a matched mode) with a duration of 2 ns, produced by a RADAN-220 generator.

Radiation in the range 200–520 nm has two main components. The first, with a short-term removal of the excitation time, can be attributed to the emission of the third continuum of xenon in the range 200–400 nm. The second component is the recombination radiation from the constricted discharge. The arrival of reflected voltage pulses at the discharge gap leads to an increase in the intensity of the third continuum and recombination radiation; however, this does not greatly affect the intensity of the second continuum.

5. Conclusion

Studies have shown that the characteristics of Ar, Xe, H_2 and N_2 emission in the VUV spectral region, when excited by a diffuse discharge formed in an inhomogeneous electric field, do not change significantly. 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.

In hydrogen, a maximum at 160 nm emission is observed in the diffuse discharge spectra and the luminescence intensity in the region of 220–280 nm is relatively low. If a little amount of argon is added into hydrogen, the uniformity of the discharge is impaired, the emission intensity of the hydrogen band with a maximum at a wavelength of 160 nm decreases and a band with a maximum at a wavelength of 191 nm appears. Emission of argon dimers in the VUV spectral region under these conditions is not detected. The electron density and excitation temperature are also diagnosed in pure argon. Experimental results indicate that a stronger electric field enhances both of the electron density and excitation temperature. Additionally, the excitation temperature decreases as the gas pressure increases.

In the case of N_2 only week VUV lines were evident, while UV emission on C-B nitrogen band dominates in the spectra.

Acknowledgements

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6. References

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