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Progress in Spectroscopic Diagnostics of Barrier Discharges

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Systematic study of barrier discharges (BDs) using cross-correlation spectroscopy (CCS) has been conducted since 2000 by two research teams at the universities in Moscow and in Greifswald. From the beginning, the investigations were focused on the application of the CCS diagnostics to the BDs in air and in N_2/O_2 mixtures. Further progress in the research field being considered was aimed on the one hand, at the improvement of the measurement technique and development of the data processing procedures, and on the other hand, at using different working gases and gas mixtures. In this paper, the most interesting and important research results obtained in the course of two last years at both referred above universities using the CCS technique are reported. They correspond to three different subjects of investigation, namely an analysis of axial and radial structure of the microdischrge in air at ambient pressure, an influence of the composition of binary mixtures Ne/O_2 upon the mechanism of the BD development, and spatio-temporal structure of the BD in pure argon.

1 Introduction

The kinetic emission spectroscopy is one of the most informative experimental methods of plasma diagnostics. However as applied to barrier discharges (BDs), this method for a long time was restricted to spatially and/or temporally integrated (averaged) measurements [1]. Certain peculiarities of the complicated spatio-temporal structure of BDs that in many cases consist of a great number of microscharges (MDs) of sub-millimetre size and nanosecond duration, make special requirements for the measurement technique. Such a technique was originally proposed in 1995 [2]. It included the method of localization of the MDs at a fixed place and the technique of cross-correlation spectroscopy (CCS, sometimes also referred to as "coincidence technique" or "time-correlated single photon counting" [3]).

Systematic studies of BDs using CCS has been conducted since 2000 by two research teams at the universities in Moscow and in Greifswald. From the beginning, the investigations were focused on the application of the CCS diagnostics to the BDs in air and in N_2/O_2 mixtures. Principal results of this work have been published in several review papers [4-6]. Further progress in the research field being considered was aimed on the one hand, at the measurement technique improvement and development of the sophisticated data processing procedures, and on the other hand, at using different working gases and gas mixtures.

In this paper, we report the most interesting and important research results obtained recently (in the course of two last years) at both referred above universities using the CCS technique. Experimental data presented below correspond to three different subjects of investigation, namely an analysis of axial and radial structure of the MD in air at ambient pressure (section 3.1), an influence of the composition of binary mixtures Ne/O_2 upon the mechanism of the BD development (section 3.2), and spatio-temporal structure of the BD in pure argon (section 3.3).

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2 Experimental set-up and procedure

The experimental set-up and the CCS-method are described in detail in [7]. Here a brief review is given. In order to localize repetitive MDs, a discharge cell consisting of two semi-spherical electrodes, both covered by dielectrics (glass or alumina) was used. This electrode arrangement provided the possibility to observe not only the volume part of the MD, but also the surface discharges (see fig. 1-a). The DBD was operated by sinusoidal voltage, at different frequencies in the range of 2-13 kHz. To investigate individual MDs, the voltage amplitude was adjusted so as to maintain the DBD in a mode corresponding to 1 MD per half cycle.

The CCS-setup is shown schematically in fig. 1, right plot. By means of a quartz lens, the discharge zone was imaged onto the optical slit. By appropriate adjustment and movement of this slit, the discharge area could be scanned in vertical (z) and horizontal (r) directions (resolution not worse than 0.1 mm). Thus localized radiation (so-called MAIN signal) was resolved spectrally by a monochromator and then detected by the high-gain photo-multiplier (PMT) operated in a single photon counting mode. The second detector (PMT) provided the synchronizing (SYNC) signal necessary for the CCS measurements (see [3,7] for the details).

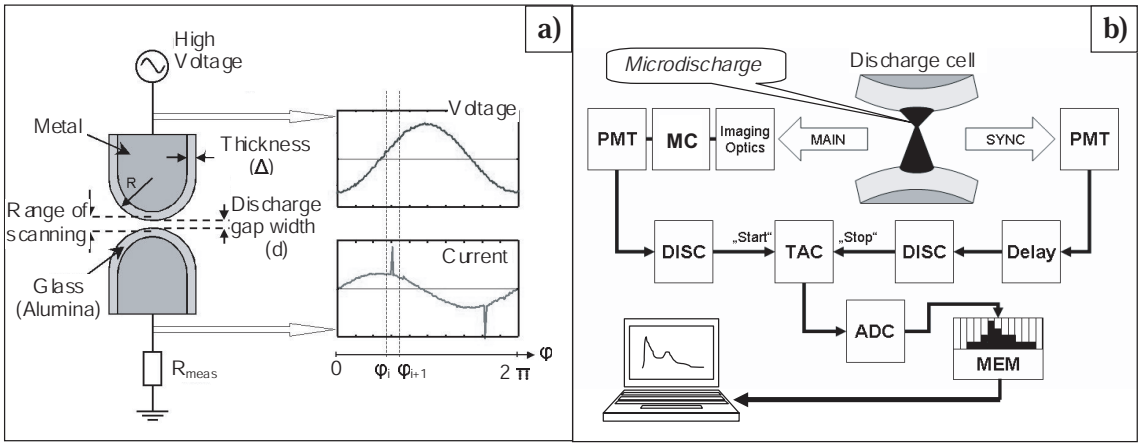

Fig. 1 General scheme of the discharge cell with indicated range of optical scanning and typical examples of the oscillograms of voltage and current (a), and a diagram of the CCS-apparatus (b). Abbreviations: PMT – photomultiplier tube, MC – monochromator, SYNC – synchronization signal, DISC – discriminator, TAC – time-to-amplitude converter, ADC – analogue-to-digital converter, MEM – memory.

The measurements of the correlation functions and accumulation of the results were carried out by the Time Correlated Single Photon Counting (TC-SPC) board. The basic components of the TC-SPC instrument are two discriminators (DISC), a time-to-amplitude converter (TAC), an analogue-to-digital converter (ADC) and a memory (MEM). Typically, up to 10^7 counts were accumulated in the memory segment with a maximal count rate for a measurement of 5 minutes of duration. The temporally resolved intensity distributions were recorded with a time resolution of about 0.1 ns (instrument resolution of 0.05 ns per channel).

3 Results and discussion

3.1 Evolution of axial and radial distributions of electric field in the MD in air.

The objective of this work was to reconstruct the evolution of axial and radial distributions of electric field within the MD of the BD in air at atmospheric pressure from experimental data on spatially 2-dimensional radiation kinetics for two selected wavelengths (λ = 337.1 nm of the 2^{nd} positive and λ = 391.5 nm of the 1^{st} negative system of nitrogen, i.e. the most intensive signals of both systems). The first attempt to realise such an idea dates back to 1999 [8]. Unfortunately, this attempt failed, since the radial distributions of the luminosities recorded under experimental conditions [8] reflected a jitter of the MD positions in their succession in radial direction rather than a real radial structure of a separate MD.

It should be noted that the first quantitative estimates of electric field strength within the MD channel from the CCS measurements of radiation kinetics for two wavelengths specified above, were accomplished within

the frame of a one-dimensional model, i.e. assuming uniform distributions of the luminosity over the radial coordinate [7]. The authors [9] succeeded to improve considerably the localisation of the MD sequences by means of an appropriate choice of the composition of the working gas mixture N_2/O_2. Their best results were achieved for the mixture containing 6 vol.% O_2 and 94 vol.% N_2. An alternative way to solve the problem of MD localisation improvement appeared to be the variation of the radii of curvature and dielectric material of the electrodes.

All experiments were performed in the discharge cell shown in fig.1 with flowing synthetic air (20% O_2 + 80% N_2) at ambient pressure. Gas flow rate was adjusted so as to avoid any accumulation of ozone in the discharge area [7]. In order to localize the BD to a definite position, we used special semi-spherical electrodes (radius of curvature R = 2 mm, see fig.1-a) covered with alumina. The discharge gap width (distance between the electrode tips) was chosen to be d = 0.9 mm. Under described above experimental conditions, the discharge demonstrated perfect localization and extremely high stability, thus allowing to carry out the time consuming spectroscopic measurements. The spatio-temporal distributions of radiation intensity for the selected wavelengths within the MD channels were recorded by means of the technique of CCS with the resolution of 0.1 mm over spatial coordinates and of 0.05 ns over time.

According to the results [10] based on the general kinetic scheme of a non-equilibrium discharge in nitrogen-oxygen mixtures, the following six elementary processes should be taken into account to describe the radiation kinetics for the second positive and first negative system of nitrogen (0-0 transitions) at atmospheric pressure.

Excitation of the molecules of nitrogen (in the ground state) by direct electron impact:

$$\mathrm{e} + \mathrm{N_2}(\mathrm{X}^1\Sigma_\mathrm{g}^+)_{\mathrm{v}=0} \;\rightarrow\; \mathrm{N_2}(\mathrm{C}^3\pi_\mathrm{u})_{\mathrm{v}'=0} + \mathrm{e} \qquad (\Delta\mathrm{E} = 11\,\mathrm{eV}) \tag{1}$$

$$\mathrm{e} + \mathrm{N_2}(\mathrm{X}^1\Sigma_\mathrm{g}^+)_{\mathrm{v}=0} \;\rightarrow\; \mathrm{N_2^+}(\mathrm{B}^2\Sigma_\mathrm{u}^+)_{\mathrm{v}'=0} + 2\mathrm{e} \qquad (\Delta\mathrm{E} = 19\,\mathrm{eV}) \tag{2}$$

Spontaneous radiation of thus formed excited species:

$$\mathrm{N_2}(\mathrm{C}^3\pi_\mathrm{u})_{\mathrm{v}'=0} \;\rightarrow\; \mathrm{N_2}(\mathrm{B}^3\pi_\mathrm{g})_{\mathrm{v}''=0} + \mathrm{h}\nu \qquad (\lambda = 337.1\,\mathrm{nm};\, \tau_\mathrm{o}^\mathrm{C} = 40\,\mathrm{ns}) \tag{3}$$

$$\mathrm{N_2^+}(\mathrm{B}^2\Sigma_\mathrm{u}^+)_{\mathrm{v}'=0} \;\rightarrow\; \mathrm{N_2^+}(\mathrm{X}^2\Sigma_\mathrm{u}^+)_{\mathrm{v}''=0} + \mathrm{h}\nu \qquad (\lambda = 391.5\,\mathrm{nm};\, \tau_\mathrm{o}^\mathrm{B} = 60\,\mathrm{ns}) \tag{4}$$

Collisional quenching of the excited species by the molecules of nitrogen and oxygen:

$$\mathrm{N_2}(\mathrm{C}^3\pi_\mathrm{u})_{\mathrm{v}'=0} + \mathrm{N_2}/\mathrm{O_2} \;\rightarrow\; \mathrm{Products} \tag{5}$$

$$\mathrm{N_2^+}(\mathrm{B}^2\Sigma_\mathrm{u}^+)_{\mathrm{v}'=0} + \mathrm{N_2}/\mathrm{O_2} \;\rightarrow\; \mathrm{Products} \tag{6}$$

Within the frame of this kinetic model, the ratio of the rate constants of the reactions (1) and (2) is a sole function of the reduced field strength: $k_2/k_1 = k_2/k_1(E/N)$, and it can be calculated from the measured radiation intensities for the selected wavelengths (see reactions (3 and (4)) at any point (z, r, t). The corresponding formulas are given in the papers [7,10].

The procedure of data processing consisted of the following basic steps: Abel inversion, numerical smoothing over time, numerical differentiation over time, evaluation of reduced field strength $E/N(z, r, t)$. Several special algorithms and computer programs were developed to realize this procedure, taking into account some peculiarities of experimental data arrays (statistical nature of the measurement results and a priori estimates of their accuracy). To evaluate reduced field strength $E/N(z, r, t)$ from the calculated rate constant ratio $k_2/k_1(z, r, t)$, we used the calibration curve $k_2/k_1(E/N)$ that was obtained experimentally [11].

An example of thus computed results is presented in fig.2. The propagation of the cathode-directed ionizing wave with the amplitude of up to 450 Td is clearly seen there. It is interesting to mention that when this ionizing wave reaches the cathode surface, it does not stop but continues its movement in radial direction from the MD axis. Therefore, the surface part of the MD on the cathode (this part of the discharge has a profound branched structure [9] which is referred to as "Lichtenberg figures" [2]) can be regarded as a natural continuation of its volume part.

It should be noted also that an estimate of the size of local field enhancement which can be derived from fig.2 to be within the range 10-100 μm, is limited in fact by the resolution of the apparatus (0.1 mm).

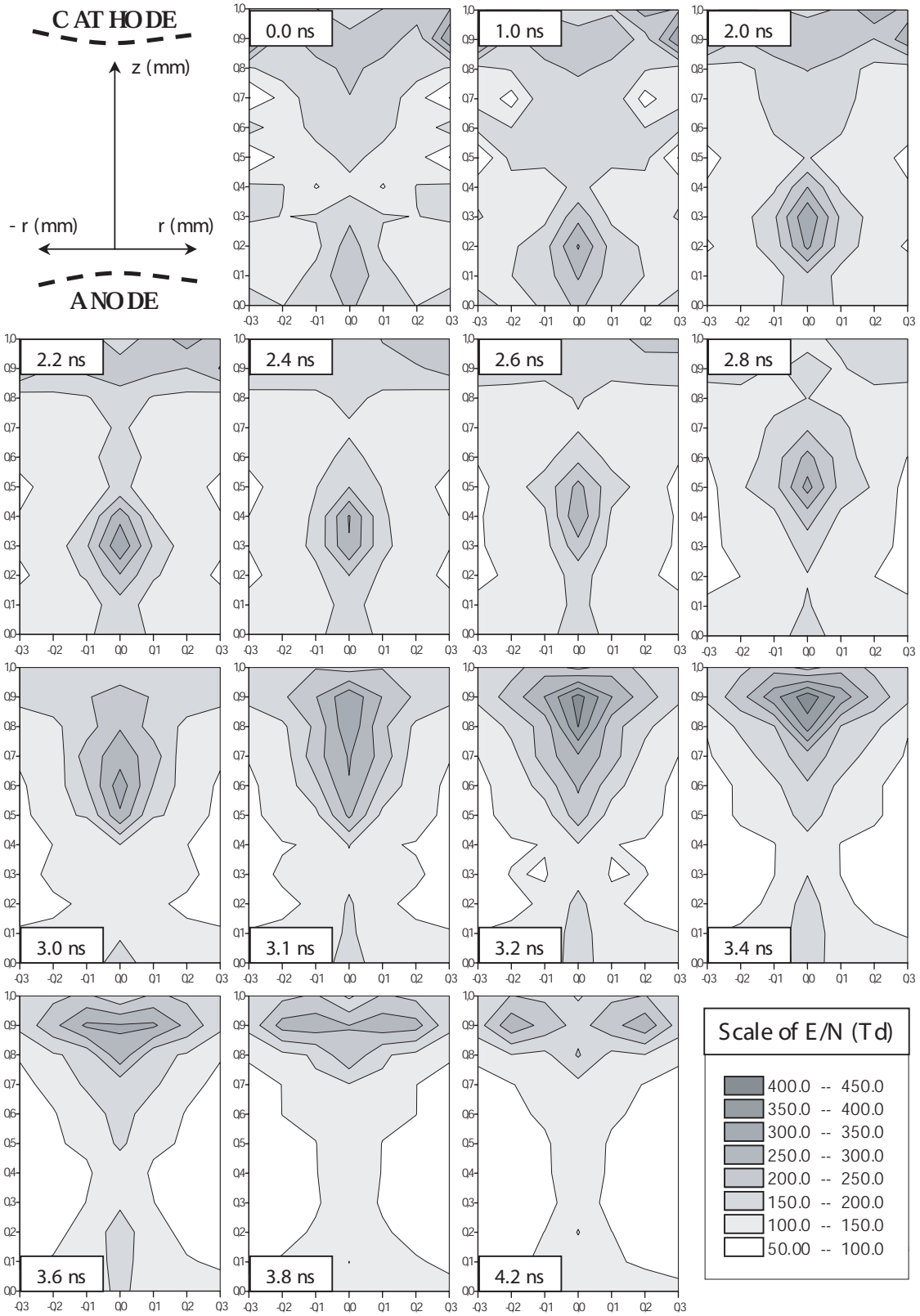

Fig. 2 Evolution of electric field within the microdischarge channel. Experimental conditions: flowing synthetic air at p = 1.035 bar; gap width (distance between the electrode tips) d = 0.9 mm; voltage frequency f = 6.03 kHz

3.2 BDs in the mixtures Ne/O_2.

In the parallel-plane electrode arrangements, the BDs in neon and in helium were demonstrated to exist in a so-called diffuse mode [12]. The latter term is used to specify a spatial distribution of the luminosity within the discharge gap which is uniform in any plane parallel to the electrode surface at any moment of the BD evolution (i.e.$I(x, y, z, t) = I(z, t)$, provided that z-axis is directed transversely to the electrode plane). A diffuse mode is also known for the BD in pure nitrogen [6,10], although its spatio-temporal structure considerably differs from those ones for neon and helium. Even small admixtures of O_2 (500-1000 ppm) to N_2 were found to cause a transition of the BD from diffuse to filamentary mode (i.e. appearance of MDs) [10]. In order to investigate the stability of the diffuse BDs in Ne and to clarify the role of molecular oxygen in the discharge mechanism, we conducted a series of CCS measurements of the spatio-temporal structures of the BD luminosities in the mixtures Ne/O_2 at ambient pressure. Oxygen concentration was varied in the range 0.1–90 vol.%.

These experiments were carried out in the discharge cell shown in fig.1-a. The semi-spherical glass electrodes with radius of curvature R = 7.5 mm were used, the gap width was equal to 1.0 mm. Feeding voltage frequency was varied in the range 2-12 kHz, but it was found to exert only a weak influence on the BD behaviour, so all the CCS measurements were conducted at 12 kHz. An influence of the O_2 content on the BD structure was clearly seen by a naked eye. In pure neon (degree of purity 99.999%), no channel between the electrodes was observed and diffuse glows near both electrode surfaces were seen. An admixture of 0.1% of oxygen resulted in the formation of a diffuse channel between the electrodes which had a diameter of a few millimetres, while the areas of the electrode glows noticeably decreased. Further increase in oxygen content caused considerable decrease in distinctly visible diameter of the MD channel, which at O_2 concentration of 1% was about 1 mm.

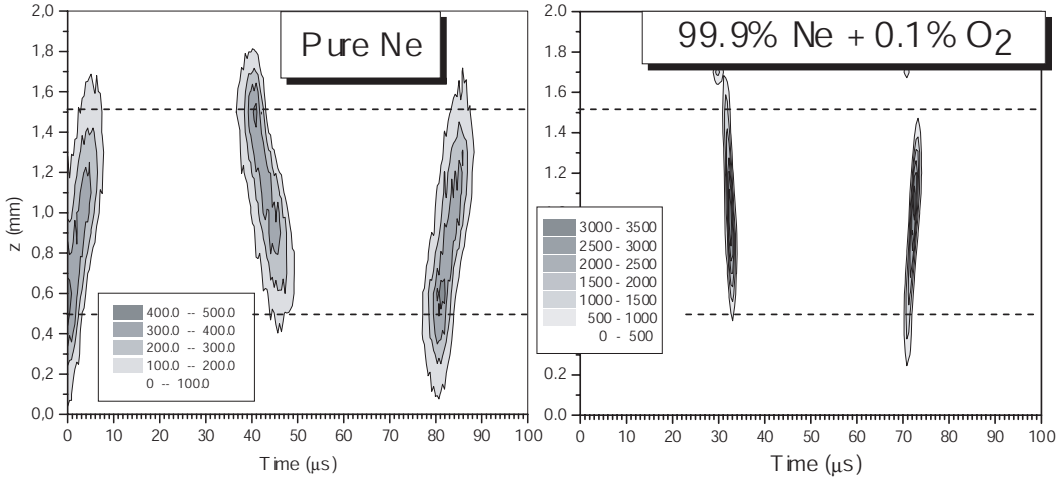

Fig. 3 Spatio-temporal distributions of the luminosity for λ = 703 nm. Experimental conditions: flowing gas (binary gas mixture) at p = 1.05 bar; the distance between the electrode tips (denoted on the figures by dashed lines) d = 1.0 mm; voltage frequency f = 12.1 kHz.

As in the case of a parallel-plane electrode arrangement [12], the spatio-temporal structure of the BD in the special discharge cell (see fig.1a) in pure neon consists of a succession of the cathode-directed waves of luminosity (see fig.3, left plot), provided that the line λ = 703 nm (Ne*, excitation energy of about 19 eV [12]) was chosen as a spectral indicator. Assuming direct electron impact to be the dominant excitation channel of this radiating state Ne* and taking into account relatively high value of excitation energy, these luminosity waves can be considered as ionization waves [7].

The comparison of the plots in fig.3 demonstrates that an increase in oxygen content results in narrowing of the luminosity waves and in considerable growth of their velocities. Such behaviour was observed for the entire studied range of oxygen concentrations. The effect of narrowing can be easily explained by collisional quenching of Ne* by molecular oxygen. An influence of oxygen content on the ionization wave velocities is determined by the dependencies of the kinetic coefficients of electrons (drift velocities, ionisation coefficients, etc.) upon the chemical composition of the binary mixtures being considered.

It seems interesting to mention that qualitatively, the axial spatio-temporal structures of the luminosities of the diffuse (pure Ne) and filamentary (Ne/O_2 mixtures) BDs are very similar. In contrast to the case of N_2/O_2 system [10], the transition between the filamentary and diffuse modes of the BD in Ne/O_2 mixtures occurs gradually.

It is also interesting to compare the mechanisms of electrical breakdown for the diffuse BDs in Ne or He (see fig.3, left plot, and the results in [12]) and for the filamentary BDs in N_2/O_2 mixtures (see fig.2 and the results presented in [4-10]). In both cases, a cathode directed wave of luminosity is observed. As already mentioned above, these luminosity waves reveal the behaviour of electric field and they can be treated as ionization waves. In the case of the diffuse BDs in Ne and He, the propagation of the cathode-directed ionizing waves lasts for a few microseconds (see fig.3, left plot), while in the case of the filamentary BDs in N_2/O_2 mixtures this process takes 1-2 nanoseconds (see fig.2).

The reasons for which the velocities of the ionizing waves are much lower in rare gases (He, Ne) than in N_2/O_2 mixtures, can be understood simply by looking at the breakdown curves $U(pd)$ of these gases [13]. Due to the very different dependence of the reduced ionization coefficient α/N as a function of E/N in rare gases and in N_2/O_2 mixtures, small overvoltage can lead to a very large increase of the electron multiplication in N_2/O_2 mixtures but is not so dramatic in He or Ne.

3.3 BD in pure argon

This experimental research was originally aimed at establishing the mechanism of the BD development in pure argon at ambient pressure [14]. However, some preliminary experiments showed that even visible shape of the discharge as well as its electrical characteristics depended strongly on the concentrations of the impurities in the ppm range. That is why an investigation of the influence of impurities upon the BD mechanism became second objective of this work.

In order to localize filamentary BD in argon to a definite position, we used special semi-spherical glass electrodes with radius of curvature R = 5 mm (see fig.1-a). The discharge gap width (distance between the electrode tips) was about 1 mm. Feeding voltage frequency was varied within the range 3-10 kHz. Under described above experimental conditions, the discharge demonstrated perfect localization and extremely high stability.

Emission spectrum of the BD in pure argon was found to consist of the peaks of Ar*, the bands of OH* radical, and the bands of molecular nitrogen (2^{nd} positive system), although the concentrations of H_2O and N_2 in argon were about 3 ppm and 10 ppm, respectively [14]. The most intensive lines corresponding to these species (λ = 727 nm for Ar*, λ=309 nm for OH*, and λ = 337 nm for N_2*,) were chosen as their spectral indicators. Spatio-temporal distributions of radiation intensity for thus selected wavelengths within the microdischarge channels were determined by means of the technique of CCS with the resolution of 0.1 mm over spatial coordinates and of 0.05 ns over time. Some results of these measurements are shown in fig.4.

The following peculiarities of the luminosity distributions presented in fig.4 should be noted. First, there are two distinct periods of the MD activity corresponding to the local maximums of radiation intensity of Ar* (40-60 ns and 300-500 ns, see upper plot in fig.4). This is an evidence for a double-step mechanism of electrical breakdown. Second, there are considerable differences in the luminosity distributions for different spectral indicators. This can be regarded as an evidence for complex radiation kinetics within the MD channel.

The phenomenon of a double-step electrical breakdown in argon was observed earlier in the BD for discharge gap width of 4 mm [15], and in a long gap for the spark breakdown [16].

The authors [16] explained the mechanism of second breakdown by the dominant role of metastables of Ar. The metastables of Ar can be also an additional (indirect) source of the radiating species OH* and N_2*. The latter assumption accounts for the complicated nature of radiation kinetics. In the case of a closed system, the properties of BD were found to change gradually. After 2-3 hours of the BD operation, the burning voltage increased, second intensity maximum disappeared, and the duration of the light pulses for all spectral indicators decreased considerably. All these changes can be explained by the effect of accumulation of impurities (quenchers) in the discharge area.

The first luminosity maximum for λ = 727 nm (Ar*) in fig.4 can be attributed to an ionization wave which propagates much faster than those ones in Ne (see fig.3). Unfortunately, we could not determine the velocity of this light front (ionization wave) due to not sufficient resolution over the time scale that was limited by certain jitter of the synchronisation signal. In contrast to the experiments in air that were conducted using a differential discriminator for synchronisation signal, the measurements in Ar were carried out on the CCS-apparatus with a

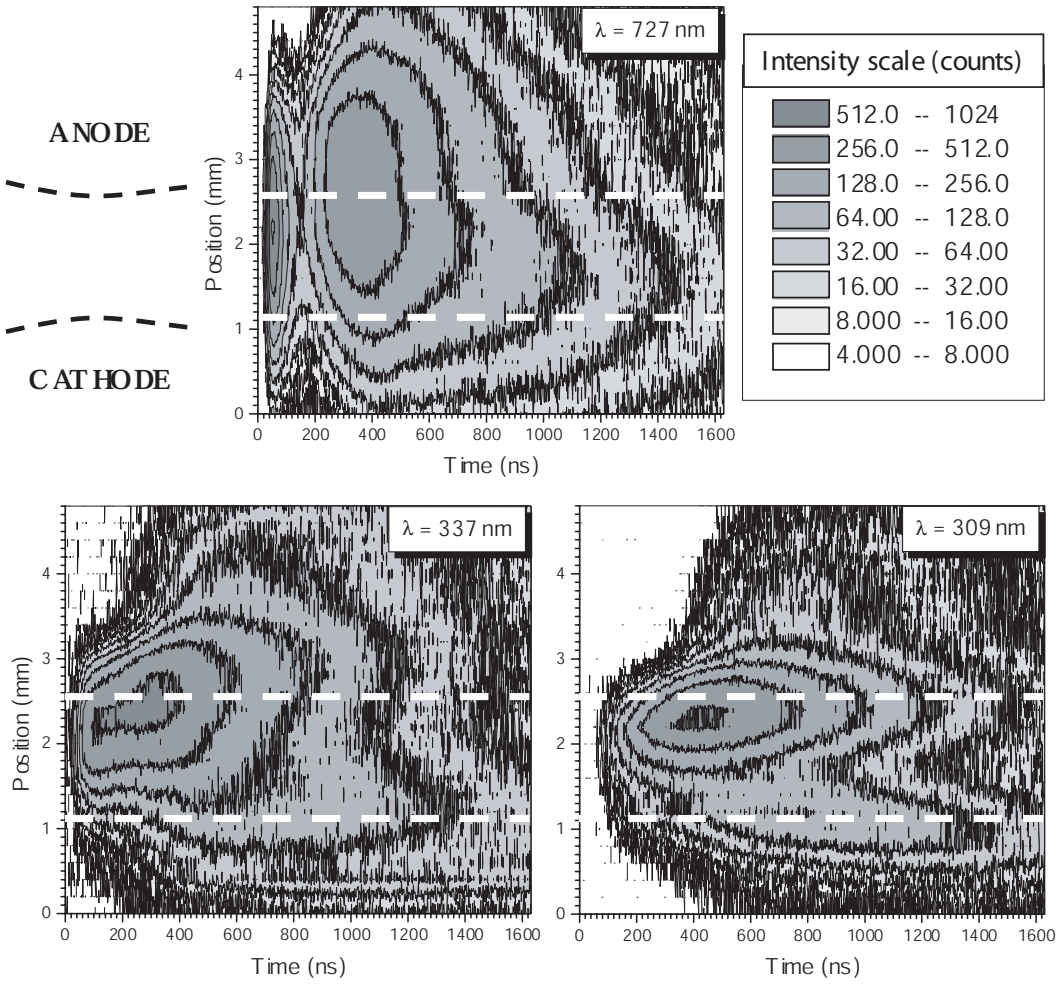

Fig. 4 Spatio-temporal distributions of the luminosity for selected spectral indicators. Experimental conditions: flowing argon at p = 1.1 bar; the distance between the electrode tips (denoted on the figures by the white dashed lines) d = 1.5 mm; voltage frequency f = 5.1 kHz.

linear discriminator which is not limited by a steepness of a signal front but provides somewhat poorer resolution over time (see [3] for the details). Our attempt to investigate the BD in pure argon by the CCS-apparatus with a differential discriminator failed (no synchronisation signal could be detected).

The degrees of purity of two rare gases that were used in the experiments described above (99.998% for argon and 99.999% for neon) are relatively high and comparable. Therefore, an influence of impurities of molecular gases that can act as quenchers of metastable states of Ar and Ne, may be excluded from the list of possible reasons for the striking difference between the mechanisms of the BDs in pure neon (diffuse mode) and the BDs in pure argon (filamentary mode, compare also the plots presented in fig.3 and fig.4). To explain this difference, the dependence of the reduced ionization coefficient α/N as a function of E/N in both rare gases should be taken into account, and then, the sensitivity of electron multiplication factor to electrical field strength must be considered [13].

4 Concluding remarks

The scope of possible applications of the CCS technique in the field of plasma diagnostics is not limited to BDs, only. It has been already used for coronas, surface discharges, and even for plasma torches. Generally, this scope can be extended to any kind of periodical or steady-state discharges.

However, the most important advance in plasma diagnostics by means of the CCS has been made for the case of the filamentary BDs. Due to extremely high sensitivity of the CCS technique and its temporal resolution in a sub-nanosecond range, the arrays of measurement results were obtained that provided the data quality (accuracy and spatio-temporal grating), sufficient for quantitative estimating of electric field and relative electron density within the MD channel. The knowledge of these basic plasma parameters provides deep understanding and enables complete theoretical description of the process of electrical breakdown, including discharge physics and plasma chemistry.

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