SPATIO-TEMPORAL RESOLVED DIAGNOSTICS OF THE SINGLE FILAMENT BARRIER DISCHARGE IN AIR

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1 Introduction

To understand the physics and chemistry under the strongly non-stationary and non-homogeneous non-equilibrium plasma conditions of dielectric barrier discharges (DBDs) in the filamentary mode, the detailed knowledge of the basic plasma parameters is necessary, especially as regards the reduced electrical field strength E/n and the properties of electrons. Although some averaged measurements of the electron density (e.g.[1]) and of the reduced field strength (e.g. [2]) have been already reported, their temporally and spatially resolved experimental determination remains a far-reaching unsolved problem. Kinetic models of the plasma chemistry under such conditions are based mainly on numerical calculations of these parameters. Experimental investigations in the filamentary mode require a time resolution in the sub-nanosecond scale and a space resolution in the sub-mm range [3]. On the base of spatially and temporally resolved measurements of single filaments by the so-called cross-correlation spectroscopy CCS [4], in the present paper this method is mainly established to contribute to the estimation of the reduced field strength.
First experimental results on the spatio-temporal development of single filaments of DBDs in dry air at atmospheric pressure are presented. The measurements allow a detailed visualisation and interpretation of the streamer development. In combination with the kinetic model [4,5] they are used to get information on the spatio-temporal development of the reduced field-strength E/n, too.

2 Experimental set-up

The spatio-temporal structure of DBD-microdischarges is studied by means of optical emission spectroscopy. A single filament is generated periodically (frequency $f \approx 6$ kHz, $U \approx 14$ kV_{pp}) between two glass electrodes with semi-spherical surfaces (test tubes: outer radius of curvature 7.5 mm, glass thickness 1.5 mm). The discharge gap width was adjusted to 1.2 mm. All the experimental results presented below were obtained in the single-filament DBD in synthetic air. The experimental set-up is shown in fig. 1. The resolution parameters are summarised in table1. A detailed description of the experimental set-up is given elsewhere [6].

Table1. Resolution parameters of the experimental apparatus

quantity	**resolution**	**device and technique**
space	$\delta x = 0.2$ mm	imaging quartz optic and stepper motor
spectrum	$\delta\lambda = 0.3$ nm	monochromator
time	$\delta t = 0.1$ ns	time-correlated single photon counting (SPC) with optical triggering (so-called Cross-Correlation Spectroscopy *CCS*)
phase (of driving sine voltage)	$\delta\varphi = 2\Pi/16$	PC pattern generator (PPG, controls memory segments of the SPC-module)

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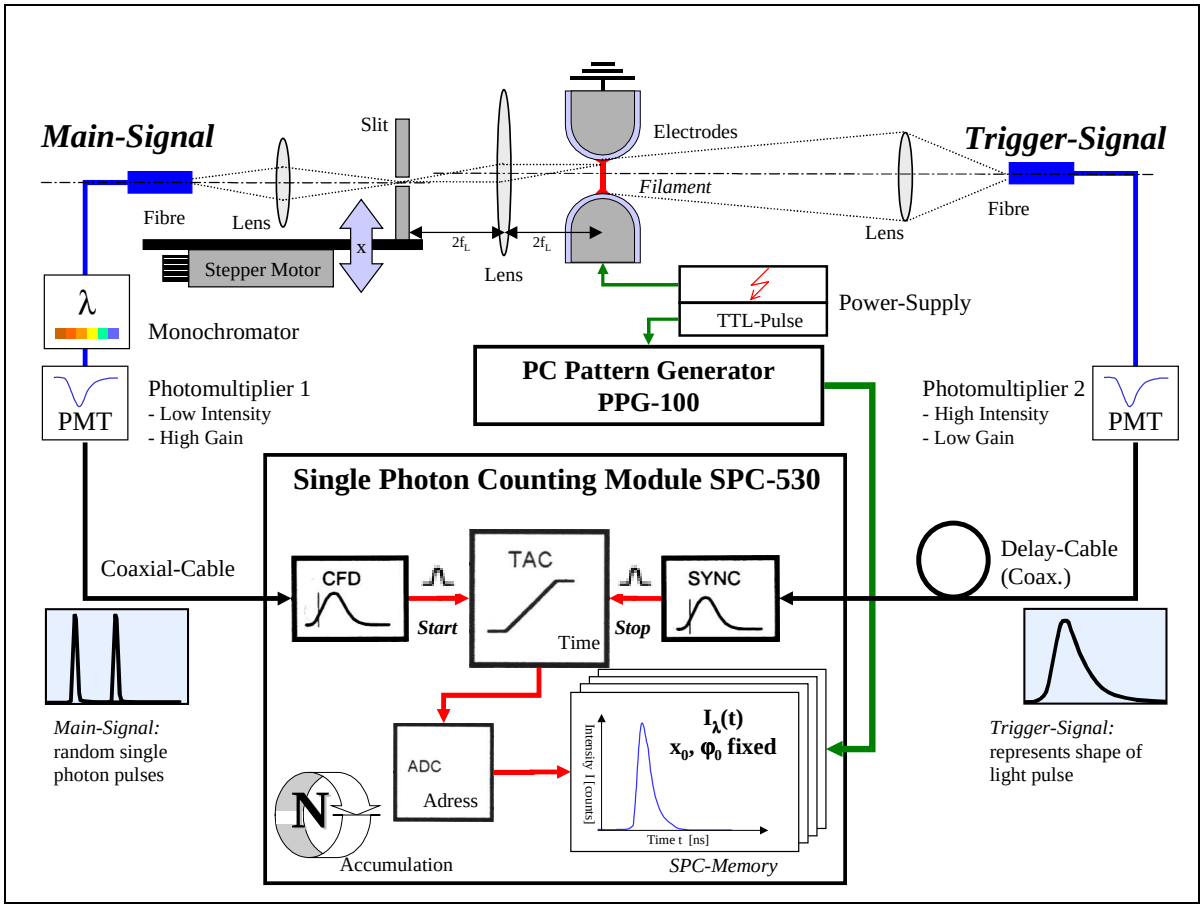

Figure 1. General scheme of experimental set-up. Abbreviations: CFD-Constant Fraction Discriminator; SYNC-Synchronisation Discriminator; TAC-Time to Amplitude Converter; ADC-Analogue to Digital Converter

3 Results and discussion

3.1 Microdischarge development

Fig. 2 shows a typical view of the PC screen for the time dependencies of light emissions of single filaments in air, resolved spectrally and by the voltage phase, after a photon collection time of 20 minutes. The statistical distribution of the filaments over the 16 PPG channels, corresponding to the phases of the applied high voltage (resolution: $\Delta\varphi = 2\Pi/16$) can be clearly seen. A noticeable photon collection could be detected only in three channels of every half period of the applied voltage.

In fig. 3 the spatially and temporally resolved intensities of the light of the first negative system (right plot) and of the second positive system (left plot), both corresponding to the phase-channel with the maximal intensity, are presented. These data agree with the experimental results [3,4] obtained under similar conditions, as regards the streamer development. The streamer propagates within the period of a few nanoseconds. It starts from the anode surface and moves towards the cathode. Then, after a relatively dark phase, it is followed by a second glow near the anode.

Qualitatively, no essential difference between the spatio-temporal distributions of luminosities for the cases of dry and humid air used as feeding gases has been observed.

3.2 Estimation of the reduced field strength (E/n)

The light emission of the first negative and second positive system of nitrogen in the DBD in air have the highest intensity. It is known from literature (e.g. [2][4, 5]), that the observed luminosity in air is determined mainly by the elementary processes summarised in table 2. It should be noted, that the stepwise processes including metastable $N_2(A)$ molecules do not play an important role in air, because of the very efficient collisional quenching of these molecules by oxygen [2, 5].

The light intensities of the corresponding spectral bands (0-0 transitions) are expected to be proportional to the local particle number densities $[N_2^+(B)]_{v=0} := n_B$ and $[N_2(C)]_{v=0} := n_C$.

For the listed in table 2 elementary processes the following system of particle balance equations can be written:

$$\frac{dn_{B,C}}{dt} = k_{B,C}\left(\frac{E}{n}\right)\cdot n_X \cdot n_e - \frac{n_{B,C}}{\tau_{0B,0C}} - \frac{n_{B,C}}{\tau_{B,C}} \approx k_{B,C}\left(\frac{E}{n}\right)\cdot n_X \cdot n_e - \frac{n_{B,C}}{\tau_{0B,0C}} \qquad (1)$$

where: k_B and k_C are rate coefficients, calculated by the integration of the corresponding cross sections and the electron energy distribution function (EEDF), the latter obtained by means of the numerical solution of the BOLTZMANN equation [2, 5],
$n_X := [N_2(X)]_{v=0}$; n_e: concentration of electrons, τ_i: time constants (comp. table 2)

Table 2. Reaction channels under consideration

First negative system of nitrogen: 0-0 transition	Rate coefficients / time constants
$e + N_2(X^1\Sigma_g^+)_{v=0} \xrightarrow{\Delta E = 19eV} N_2^+(B^2\Sigma_u^+)_{v'=0} + 2e$	$k_B(E/n)$
$N_2^+(B^2\Sigma_u^+)_{v'=0} \xrightarrow{\lambda = 391nm} N_2^+(X^2\Sigma_g^+)_{v=0} + h\upsilon$	$\tau_B \approx 60ns$
$N_2^+(B^2\Sigma_u^+)_{v'=0} + N_2 / O_2 \rightarrow N_2^+(X^2\Sigma_g^+)_{v=0}$	$\tau_{0B} \approx 0.07ns$
Second positive system: 0-0 transition	
$e + N_2(X^1\Sigma_g^+)_{v=0} \xrightarrow{\Delta E = 11eV} N_2(C^3\Pi_u)_{v'=0} + e$	$k_C(E/n)$
$N_2(C^3\Pi_u)_{v'=0} \xrightarrow{\lambda = 337nm} N_2(B^3\Pi_g)_{v=0} + h\upsilon$	$\tau_C \approx 40ns$
$N_2(C^3\Pi_u)_{v'=0} + O_2 \rightarrow N_2(X^1\Sigma_g^+)_{v=0} + O + O$	$\tau_{0C} = 0.5 \ldots 0.8ns$

The particle number densities are correlated with the counting rates, hence with the total numbers of counts for a chosen collection time ($t_{collection}$) of photons by:

counting rates $i(B,C)$: total number of counts $I(B, C)$:

$$i(B) = C_B \cdot n_B / \tau_B \qquad I(B) = i(B) \cdot t_{collection}$$
$$i(C) = C_C \cdot n_C / \tau_C \qquad I(C) = i(C) \cdot t_{collection} \qquad (2)$$

where C_i are the corresponding apparatus constants.
The combination of the systems (1) and (2) results in the following equation for the ratio of the excitation coefficients, the number of collected counts, their temporal derivation and the corresponding time constants:

$$\frac{k_B}{k_C}\left(\frac{E}{n}\right) = C \cdot \left(\frac{\tau_B}{\tau_C}\right) \cdot \frac{\dot{I}_B + I_B / \tau_{0B}}{\dot{I}_C + I_C / \tau_{0C}} \qquad (3)$$

Here C is a generalised constant which may be estimated by calibration
The ratio k_B / k_C (E/n) has been already calculated by means of the numerical solutions of the BOLTZMANN equation for the weakly ionized plasma in air under the quasi-stationary conditions [2, 5]. These results are shown in fig. 4, too. Assuming the validity of the local equilibrium concept, (i.e. that the EEDF follows any variation of the reduced field strength within a time scale, less than that one of the streamer development), these results were used to estimate the spatio-temporal structure of (E/n) from our experimental results by means of equation (3). A typical example of such an estimation is given in fig. 5 (calibration constant $C=0.01$). As it has been expected, the maximum value of the reduced field strength appears to follow the development of the streamer. Any C-value variation has been found to exert certain influence on the maximum value of (E/n), rather than on the qualitative picture of the (E/n) distribution.
At present the experiments are in progress, aimed to get an accurate calibration of the dependence (3) using space charge free plasmas.

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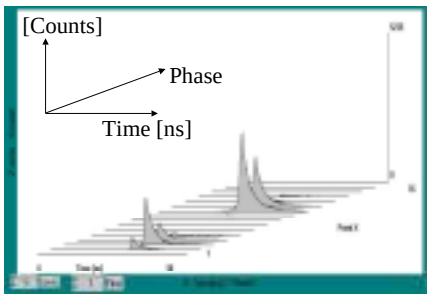

Figure 2.
Typical view of the PC screen, illustrating the temporally and phase resolved development of single microdischarges (second positive system; 0-0 transition; λ= 337 nm) by means of the SPC-530. Voltage phase resolution: Δφ = 2Π/16;
collection time of photons: 20 minutes.

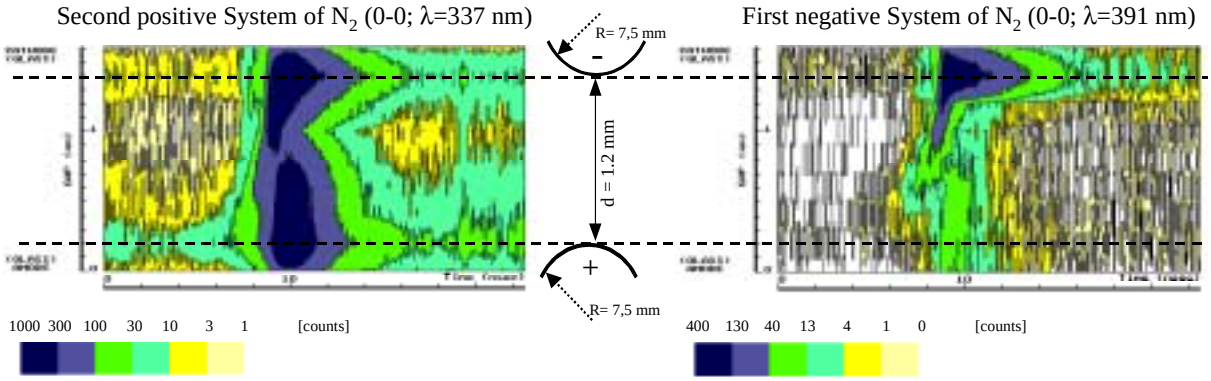

Figure 3. Spatially and temporally resolved luminosity of the microdischarge in dry air

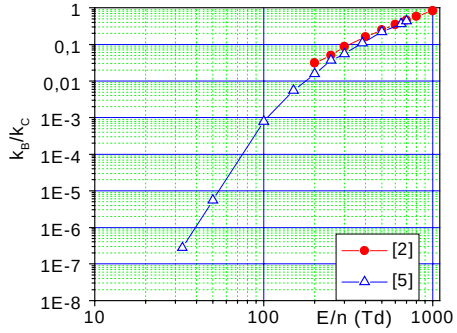

Figure 4. Calculated ratios k_B/k_C versus the reduced field strength. The solutions of the BOLTZMANN equation [2, 5].

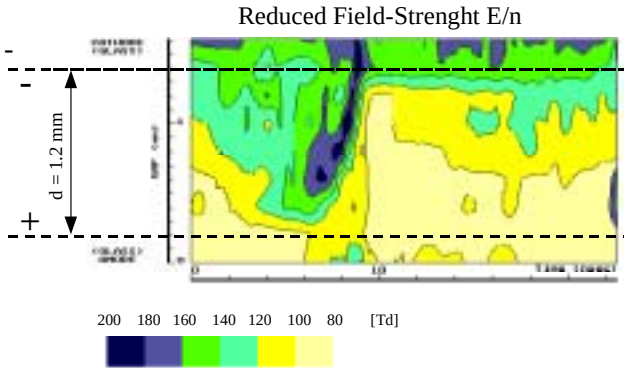

Figure 5. The evolution of (E/n), estimated on the base of results in figs. 3 and 4 as well as the kinetic model described above (C=0.01).

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