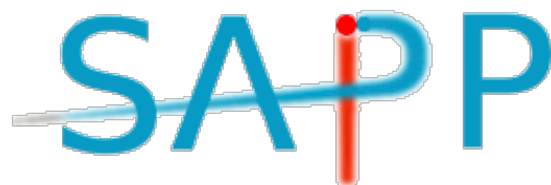


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MEASUREMENT OF THE ELECTRON DENSITY IN TRANSIENT SPARK DISCHARGE

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The paper presents a study of transient spark: a streamer-to-spark transition discharge in air at atmospheric pressure. The transient spark (TS) is applicable for flue gas cleaning or bio-decontamination and has a potential in combustion, and flow control applications. Despite the DC applied voltage, TS has a pulsed character with short (~ 10 - 100 ns) high current (>1 A) pulses, with repetitive frequencies 1-10 kHz. The electron density $n_e \sim 10^{17}$ cm⁻³ at maximum is reached in TS using relatively low power delivered to the plasma (0.2-3W).

The estimate of temporal evolution of n_e was derived from the resistance of plasma, obtained by detailed analysis of the electric circuit representing TS and the plasma diameter measurements using a fast iCCD camera. This estimate was compared with n_e calculated from the measured Stark broadening of H α line. Good agreement was obtained when plasma diameter was approximated using the full width at the half maximum of the radial emission intensity profile of the plasma channel after the Abel inversion.

1. Introduction

Various electrical discharges were found to be useful for the generation of highly reactive plasma. Plasma can initiate chemical reactions in otherwise inert gaseous mixtures. Many potential applications of plasma utilization were thus already tested, including bio-decontamination of water, surface processing, exhaust gas control of plasma assisted combustion. Because of the broad span of possible applications with different demands on plasma properties, there is no single universally useful type of electrical discharge. New discharges are therefore still being developed and studied.

We study a filamentary discharge of streamer-to-spark transition type named transient spark (TS). TS is a relatively new type of discharge, it was first mentioned in [1] and further described in [2], yet its concept is similar to the prevented spark studied in the group of Marode [3-5]. Despite the DC applied voltage, TS has a pulsed character with short (~ 10 - 100 ns) high current (>1 A) pulses, with repetitive frequencies 1-10 kHz [6].

Thanks to the short current pulse duration, the plasma generated by TS has different properties than the plasma generated by an ordinary spark. Our previous time-integrated optical emission spectroscopy (OES) study showed that TS pulses generate highly reactive non-equilibrium plasma with excited atomic radicals (O*, N*), excited molecules N₂* and ions N₂⁺, and gas temperature T_g ranging from 500 to 1500 K, while vibrational temperature T_v was from about 3800 K to 5000 K [7].

The OES is one of the most common non-invasive plasma diagnostic methods. The OES technique can provide valuable information on plasma properties [8, 9]. Besides the determination of the rotational, vibrational, and electronic excitation temperatures of excited plasma species, it can be also used to calculate the density of electrons from Stark broadening of atomic emission lines. The electron density n_e is crucial parameter for the assessment of the plasma reactivity, since the whole plasma chemistry is initially induced by high energy electrons.

There is also an alternative method for the calculation of n_e . Density of electrons can be derived from the conductivity and dimensions of the plasma. In this paper we used this approach for the calculation of time evolution of n_e in TS discharge. The plasma conductivity was calculated using oscilloscopic measurements, while the plasma dimensions were obtained by time-resolved imaging using high-speed iCCD camera. Results were compared with n_e calculated from the Stark broadening of H α emission line.

2. Experimental setup

Figure 1 shows a simplified scheme of the experimental set-up. Experiments were carried out at room temperature in atmospheric pressure air with a gas flow perpendicular to discharge channel with a velocity of $\sim 0.2 \text{ m}\cdot\text{s}^{-1}$. A stainless-steel needle was used as a HV electrode opposite a grounded planar copper electrode. The distance between electrodes d varied from 4 to 6 mm.

A dc high voltage (HV) power supply connected via a series resistor R limiting the total current was used to generate a positive TS discharge. The value of R varied from 3.5 to 9.84 M Ω . An additional small resistor $r = 1 \text{ k}\Omega$ was attached directly to the HV electrode, separating it thus from a HV cable connecting it with the resistor R (figure 1). The role of r was to eliminate the oscillations of electric signals caused by internal inductances of the HV cable and of a grounding wire.

The discharge voltage was measured by two 100 M Ω HV probes (Tektronix P6015A) at both ends of the resistor r . The currents at the grounded electrode and between R and r were measured by a Pearson Electronics 2877 (1V/A) and a Pearson Electronics 4100 (1V/A) current probes. All current and HV probes were linked to the 200 MHz digitizing oscilloscope Tektronix TDS2024 with a sampling rate up to 2 Gs s^{-1} .

Images of single TS pulses were taken by an intensified CCD camera (Andor Istar) with a time resolution down to 2 ns. The iCCD camera was triggered by a generator of 5 V rectangular pulses, which was triggered directly by the current signal. For this purpose we measured the discharge current also on a 50 Ω or 1 Ω resistor shunt. This enabled us to synchronize the acquisition of the emission either with the beginning of the streamer or with the beginning of the spark, depending on the resistor shunt used. However, in both cases we are unable to acquire the initial 25 ns of emission due to a delay caused by the trigger generator, transmission time of the signal by BNC cables and a camera insertion delay.

The time-resolved emission spectra were obtained using the same iCCD camera coupled to a 2 m monochromator Carl Zeiss Jena PGS2 (resolving power 45000), covering UV and VIS regions (200-800 nm). Here the delay between the current signal and opening of the camera was compensated using a 10 m long optical cable (Ocean Optics P400-10-UV-VIS).

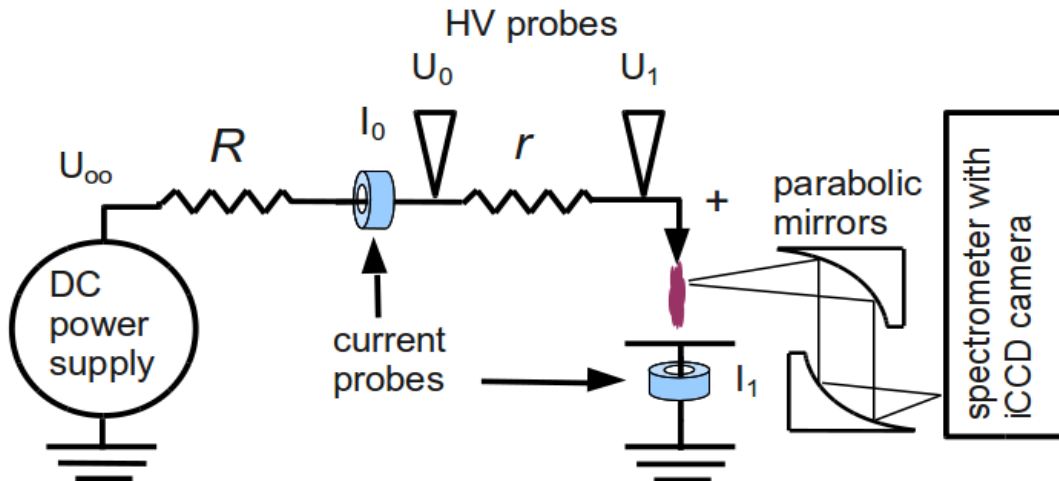


Fig. 1. Simplified scheme of the experimental set-up.

3. Results and Discussion

When the high voltage U_{00} applied to the stressed electrode is progressively increased, we first observe a streamer corona. When the breakdown voltage is reached, a transition to transient spark occurs at the discharge voltage U_{TS} . TS is a filamentary streamer-to-spark transition discharge initiated by a streamer, which transforms to a short spark pulse (phase B, Fig. 2). The duration of streamer to spark transition period (phase A, Fig. 2) is from around 100 ns to several μs , depending on TS repetition frequency [10].

The TS current pulse is due to the discharging of the capacity C , composed of several components (internal capacity of the discharge chamber C_{int} , capacity of the high voltage cable C_{cable} between the

ballast resistor R and the electrode, and capacity of the HV probe $C_{HV} = 3$ pF). When C is discharged, the current approximately given by

$$I_1(t) \approx -C \frac{dU_1(t)}{dt} \quad (1)$$

reaches a high value (~ 1 A) and the voltage drops to almost zero (phase C, Fig. 2). Then, during the quenched phase, C is recharged by a growing potential U_I on the stressed electrode. As soon as U_I reaches the characteristic TS breakdown voltage U_{TS} , a new TS pulse appears. TS is thus based on charging and re-charging of capacity C . A characteristic repetition frequency f of this process is in order of several kHz, and it can be controlled by the generator voltage U_{00} . The control of TS by U_{00} and other external circuit parameters was described in detail recently [6]. More accurate version of equation (1), which takes into account the influence of r , was also provided therein.

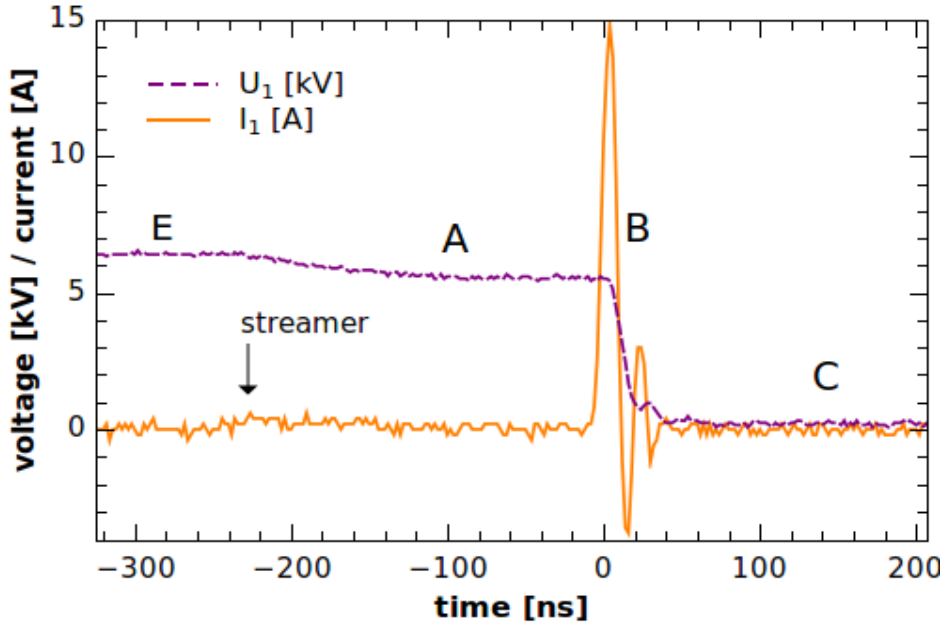


Fig. 2. Typical waveforms of transient spark in ns time scale, $R = 6.5$ M Ω , $f \approx 2$ kHz, $r = 0$ Ω , $C = 43 \pm 4$ pF, $d = 5$ mm.

Analysis of an electric circuit representing TS described in [6] enabled us to derive the plasma resistance R_p from the measured values of U_0 , U_I and I_I . From R_p , we can estimate the plasma conductivity σ_p and the electron density using following equation:

$$n_e = \frac{\sigma_p m_e v_c}{e^2}. \quad (2)$$

Here e and m_e are the electron charge and mass, respectively, and v_c is the electron-heavy particles collision frequency. The plasma conductivity σ_p is related to the plasma resistance R_p by

$$\sigma_p = \frac{d}{R_p A}, \quad (3)$$

where d and A are the gap length and the cross-sectional area of the plasma channel, respectively.

In addition to the uncertainty of R_p , the plasma diameter D_p , necessary to calculate A and v_c are major sources of uncertainty in the estimation of n_e . We calculated v_c in air for T_g from 300 to 3000 K and for reduced electric field strength E/N from 10 to 200 Td using our package for Monte Carlo simulation of electron dynamics [11]. We found that within this region of T_g and E/N , the value of v_c can vary from approximately 2×10^{11} to 4×10^{12} s $^{-1}$. Since both T_g and E/N certainly changes during the evolution of TS, v_c must change as well. We can relatively well describe the time evolution of T_g during the TS discharge but we have not data for the evolution of E/N . However, we took the calculations of Naidis [12] as sufficient approximation of E/N evolution during various phases of the TS discharge. Based on

these data, so we decided to use the constant value 10^{12} s^{-1} for v_c during all phases of TS. This approximation introduces the uncertainty of less than a factor of 4.

In order to estimate D_p , we assume that plasma diameter can be well defined by the volume from which emission can be observed. We therefore performed a set of experiments of TS imaging by iCCD camera (see Fig. 3 for illustration). We defined the exposure time and triggered the event to see either the emission from a whole single TS pulse (streamer + spark phase), or the emission of the streamer phase only, or the emission from the spark phase only. When we triggered the iCCD camera by the streamer current (measured on 50Ω shunt) we imaged the whole pulse (exposure time $0.5\text{-}2 \mu\text{s}$) or the streamer only (exposure time $25\text{-}150 \text{ ns}$). When we triggered by the spark current (measured on 1Ω shunt) we imaged the spark phase only (exposure time $25\text{-}500 \text{ ns}$).

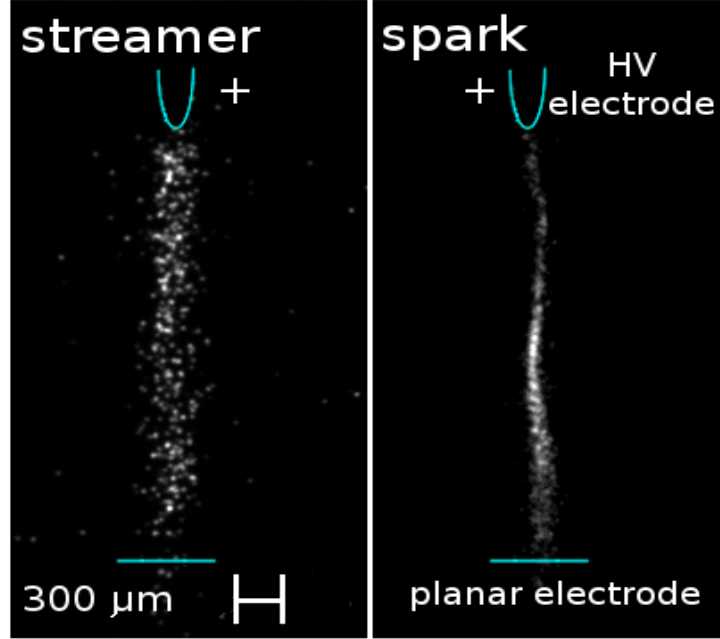


Fig. 3. Images of a single pulse in different phase of TS taken by iCCD camera; $r = 0.9 \text{ k}\Omega$, $R = 6.84 \text{ M}\Omega$, $C = 32 \pm 4 \text{ pF}$, $d = 4 \text{ mm}$, $f \approx 2 \text{ kHz}$.

In the first approximation we used the thickness of the illuminated area in the image as the plasma diameter. At lower TS repetition frequencies (below 4 kHz), we see the shrink of the plasma channel diameter during the streamer-to-spark transition from $\sim 300 \mu\text{m}$ down to less than $\sim 100 \mu\text{m}$. This is in agreement with calculations of Naidis [12]. For the estimation of n_e , we initially decided to use an average value $200 \mu\text{m}$ for all phases of TS discharge.

The estimated electron density during the spark phase of TS was found to be $\sim 10^{16} \text{ cm}^{-3}$ [6, 13]. This is high enough to cause Stark broadening of atomic lines of hydrogen. We therefore performed a preliminary study of Stark broadening of H_α line (656.28 nm), using humid air. The input air was simply bubbled through a vessel with water. We did not quantitatively measure the achieved humidity, but it was low that we observed no significant influence on electric parameters of TS. For this reason, the intensity of observed emission was quite weak, since we measured with exposure time only 20 ns .

Next, we also had to use H_α line instead of H_β line, which was too weak. Calculation of n_e from full width at the half maximum (FWHM) of H_α line is difficult due to the sensitivity on electron temperature and ion dynamics. However, Gigosos et al. [14] showed, that we can avoid these difficulties using full width at the half area (FWHA) instead of FWHM and to calculate n_e by formula:

$$\Delta\lambda_{FWHA}^{H_\alpha} = 0.549 \text{ nm} \times \left(\frac{n_e}{10^{23} \text{ m}^{-3}} \right)^{0.67965}. \quad (4)$$

Here, $\Delta\lambda_{FWHA}^{H_\alpha}$ is the FWHA of the H_α line in nm. In order to obtain reasonable values of n_e , the FWHA must be calculated from the line profile corrected with respect to Doppler, pressure and instrumental broadening. However, the minimum value of FWHM we experimentally observed was $0.14 \pm 0.03 \text{ nm}$ (time $\sim 150 \text{ ns}$ after the peak of the spark current pulse). This value is so large that

Doppler and pressure broadening can be neglected. Even after the deconvolution of the measured line profile with the slit function describing instrumental broadening, the value of this FWHM decreased to ~ 0.12 nm. Thus, the influence of instrumental broadening is within the experimental uncertainty of the measured value of FWHM. In the spectra obtained in time windows closer to the current peak where Stark broadening is even stronger, the instrumental broadening can be therefore completely neglected. Figure 4 shows normalized H_α line profiles measured in different times after the peak of the spark pulse.

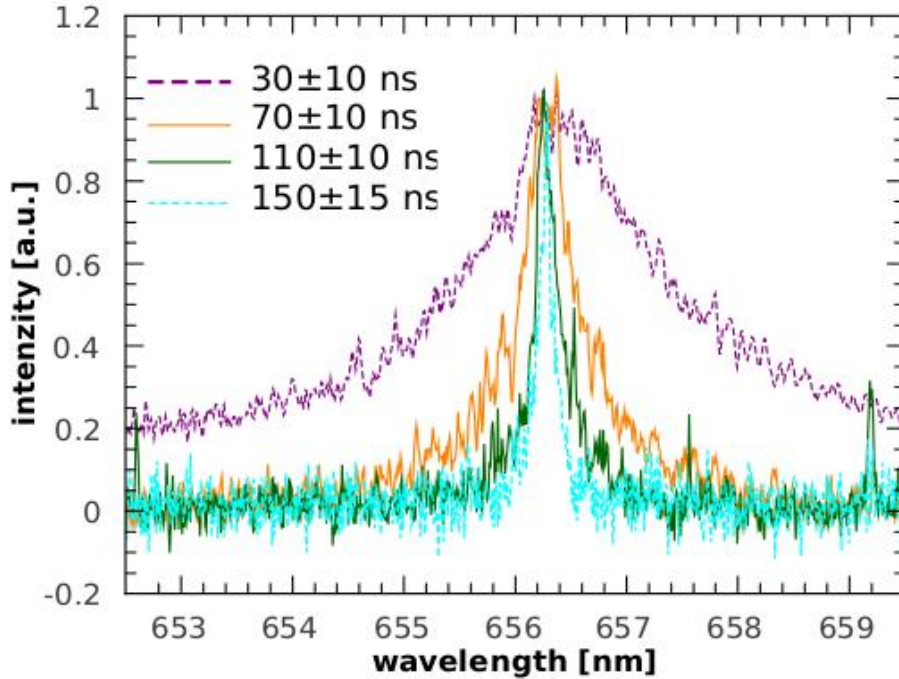


Fig. 4. Normalized emission profiles of H_α line at different times after the beginning of the spark.

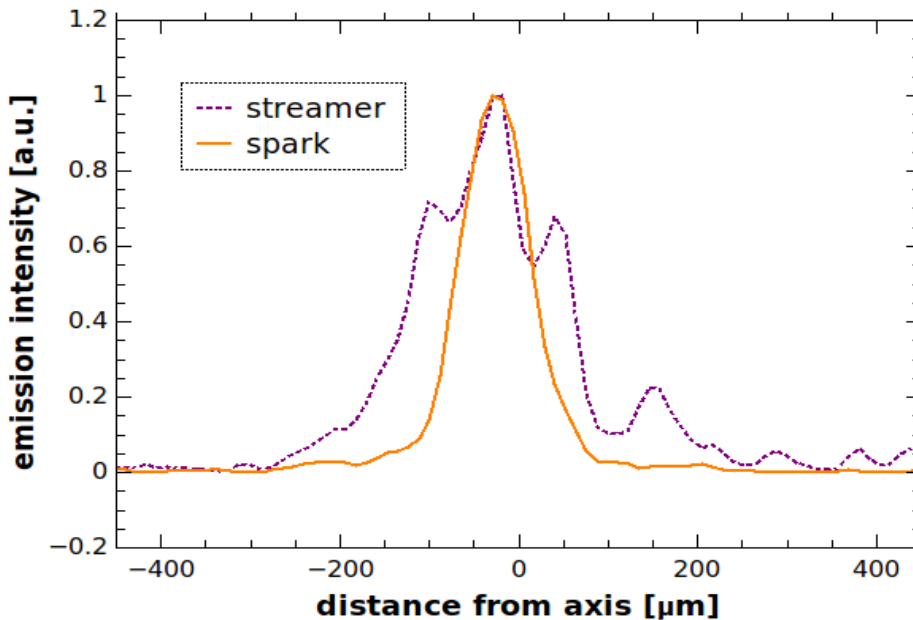


Fig. 5. Radial intensity profiles of single streamer and spark pulses

Values of n_e calculated from the measured Stark broadening were compared with estimated values and it was found that measured n_e is at least one order of magnitude higher than the initial estimate [13]. This disagreement can be resolved by more accurate estimate of the plasma diameter. More accurately,

we use the FWHM of the radial intensity profile of the plasma channel as D_p (Figure 5). After the Abel inversion, we found that the diameter of the streamer is $\sim 155 \mu\text{m}$. This agrees with the results of Gibert, et al. [15] or Van Veldhuizen, et al. [16] who also applied optical emission measurement. However, it is much larger than the streamer diameter $40 \mu\text{m}$ measured by Bastien, et al. [17] who measured it from the broadening of H lines that is related to the current density, so it may be closer to real plasma “electrical” diameter. The subsequent TS channel was found narrower (at least for $f < 4 \text{ kHz}$): less than $100 \mu\text{m}$ (FWHM after the Abel inversion being $\sim 55 \mu\text{m}$). It is slightly expanded close to the planar electrode, but we used the value typical for the region near the needle electrode, where we also measured emission spectra of H_α line. Finally, the measured n_e from broadening of H_α line is now in a good agreement with the estimate of n_e from the plasma resistance (Fig. 6).

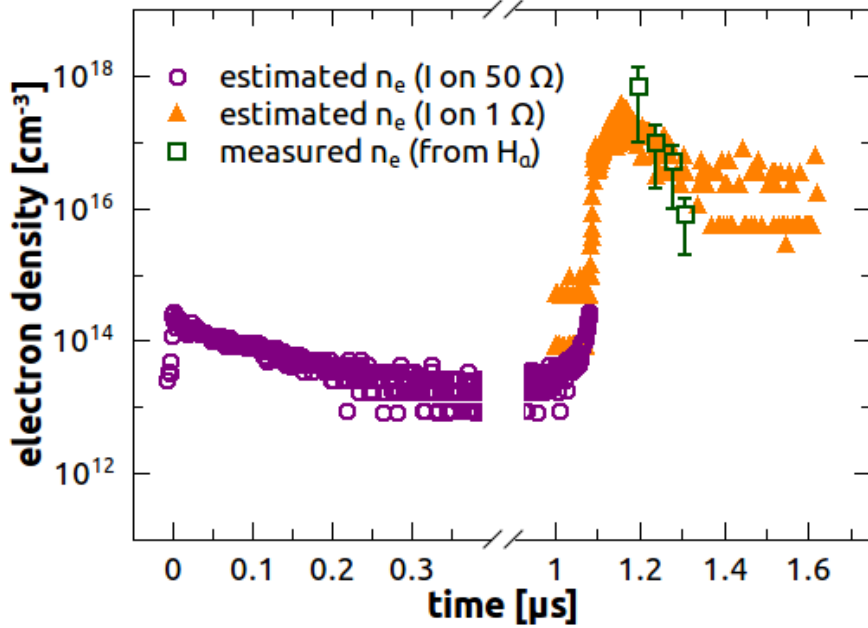


Fig. 6. Electron density during a streamer and TS pulse in μs time scale, calculated and measured by H_α broadening, $f \approx 2 \text{ kHz}$ and $C = 32 \pm 4 \text{ pF}$.

4. Conclusion

A relatively new concept of a DC-driven self-pulsing discharge was investigated: transient spark, a repetitive streamer-to-spark transition discharge of very short pulse duration ($\sim 10\text{-}100 \text{ ns}$) and with a very limited energy so that the generated plasma is highly non-equilibrium. This discharge can be maintained at low energy conditions (up to 1 mJ/pulse) by an appropriate choice of the resistances and capacities in the electrical circuit. Its frequency can be controlled by the applied voltage. The activity of transient spark is comparable with the nanosecond repetitive pulsed discharges but its advantage is an ease of the DC operation and no need of special and expensive high voltage pulsers with high repetitive frequency and nanosecond rise-times.

A fast iCCD camera was used to image the discharge diameter D_p during the streamer and the spark phase. The knowledge of plasma diameter along with a detailed analysis of the electrical circuit representing TS enabled us to estimate the temporal evolution of electron density from the plasma resistance. In order to verify this estimate we performed a preliminary measurement of time-resolved n_e from H_α Stark broadening using the fast iCCD camera coupled to a monochromator. The Stark broadening indicate n_e as high as 10^{17} cm^{-3} in the spark phase of TS at repetition frequency $\sim 2 \text{ kHz}$.

A good agreement of n_e estimated from the plasma resistance with n_e measured by Stark broadening was possible only after better approximation of D_p , using the full width at the half maximum of the radial intensity profile of the plasma channel after the Abel inversion. Different values of plasma diameter had to be used for the streamer ($\sim 155 \mu\text{m}$) and spark ($\sim 55 \mu\text{m}$) phases of TS discharge.

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