Optical diagnostics of streamers: from laboratory micro-scale to upper-atmospheric large-scale discharges

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Abstract. Optical emission produced by streamers is determined by spatial distribution of electronically excited atomic and diatomic species within the streamer head and streamer channel. Peculiarities of emission and LIF diagnostics dedicated to investigating the basic structure of streamers with high spatio-temporal resolution are discussed. Possible strategies based on the 2D projections of cylindrically symmetric streamers to determine radial distributions of excited species within the streamer channel are illustrated for streamers produced in volume or on the dielectric surface at atmospheric and low pressures.

1. Introduction

A streamer is a common form of transient discharge which develops from an electron avalanche in an overvolted gap. Streamers can be produced in the gas phase between metallic electrodes, between electrodes covered (fully or partially) with a dielectric barrier, on the interface between gaseous and liquid phases and in liquids [1]. Various atmospheric-pressure streamer-based discharges have become important in numerous environmental applications, material treatment technologies and aerodynamic and biomedical applications. Therefore continuous effort is required in order to understand and determine basic streamer parameters produced under various conditions through the state-of-the-art experimental measurements and numerical modelling.

On the other hand, red-sprite transient luminous events (TLEs) are large streamer events propagating vertically in the upper atmosphere surpassing altitudes from ~35 km to ~85 km above sea level characterised by very different pressures (between ~8.9 and ~0.16 torr) and gas temperatures (between -10 and -75 °C). Investigation of TLEs based on plasma-induced emission is, compared with laboratory discharges, much more difficult because of their stochastic nature (as in the case of cloud-to-ground lightning events) and very long observational distances (typically hundreds of kilometres between the place of the TLE occurrence and diagnostic instrumentation). Consequently, emission spectra of red sprites are acquired with low spectral, spatial and temporal resolutions. In addition, all optical signals are registered with low signal-to-noise ratio, and therefore their use in advanced
analysis which might provide estimation of important parameters (E/N, temperatures/distributions of excited species) is very limited. To this end, laboratory red-sprite TLE analogue experiments are important for better understanding of both the fundamentals of TLEs and their associated phenomena (heating or NOx production) [2].

The most important characteristics of filamentary streamers are their relatively high propagation velocity (exceeding the velocity of electron drift), small streamer channel radius (with respect to length) and enhanced mean energy and density of free electrons in the streamer head (compared with the streamer channel). All these characteristics make streamer discharges extremely difficult for observations with adequate spatio-temporal resolutions.

2. Streamer-induced emission

Optical emission produced by streamers (either at high pressure or under TLE conditions) is determined by the spatial distribution of various species within the streamer channel excited to radiative states by streamer head electrons [2, 3]. Radial distributions produced by streamer electrons are strongly non-uniform because the maximum electric field of a propagating streamer occurs on the axis of the streamer and rate constants of the excitation, dissociation and ionization processes exponentially depend on the E/N. Radial non-uniformity of excited species has to be considered in the use of diagnostics with limited spatial and temporal resolutions or in comparison of experimental results with numerical simulations [3]. In the case of nitrogen/air streamers, the \((B^3Π_u \rightarrow A^3Σ_u^+)\) first positive (FPS) and \((C^3Π_u \rightarrow B^3Π_g)\) second positive (SPS) of \(N_2\), as well as the \((B^2Σ_u^+ \rightarrow X^2Σ_g^+)\) first negative system (FNS) of \(N_2^+\), are the main components of radiation emitted in the UV-vis-NIR spectral region which can be utilized for emission-based diagnostics.

3. Optical diagnostics

3.1. Timing issues

Recording the instantaneous development of streamer emission is quite a challenging task because of the high velocity of the streamer propagation. The key issue is managing the synchronization of the discharge event with a light-detecting device(s) using a suitable triggering procedure, usually with sub-nanosecond temporal precision. In the case of pulsed or pulse-triggered discharges the problem can be solved by using a digital delay generator, which starts gating the ICCD or recording the photomultiplier (PMT) signal at the right moment. The discharge event is externally triggered on request as is the detector.

In the case of streamer discharges with temporally unstable appearance (e.g. Trichel pulse corona, barrier discharges driven by AC waveforms or temporally irreproducible pulsed discharges with low-gradient voltage slopes) no well-defined external trigger is available. The only solution is to trigger the detecting devices with the discharge itself, utilizing particular discharge events complemented by a system of delay lines. For example, current pulse produced by the discharge event in an external circuit or discharge-induced light pulse can provide precise timing of a just-past event. However, in order to compensate for the dead time (usually tens of nanoseconds) between the trigger input and the onset of the ICCD’s intensifier, the light of the discharge which is to be detected must be delayed before arrival at the detector (for example, by a sufficiently long optical fibre bundle). A similar but technically more demanding option, the time-correlated single photon counting-based technique, is discussed in Section 3.2.

3.2. TCSPC- and PMT-based scanning systems

By adjusting the electro-optical set-up of the system to detect rare single photons and to record the delayed synchronization signal from the same discharge event one can take advantage of delay-based methods and apply advanced systems which measure the probability-density functions representing the discharge emission under given conditions. The so-called time-correlated single photon counting technique (TCSPC, also known as cross-correlation spectroscopy, CCS [4,5]) is based on this
principle. For selected spatial coordinates, the TCSPC method substitutes for the real-time emission measurement of the discharge event with statistically averaged determination of cross-correlation function between two optical signals originating from the same source. These are the so-called ‘main signal’ (the spatially and spectrally resolved single photon, the actual emission detection) and the ‘synchronizing signal’ (the integral light intensity pulse of the discharge event which gives the time reference). The time between the detections of these signals is measured. Consequently, time histograms of counted photons for all spatial positions of the discharge are accumulated.

Figure 1 shows an example of a TCSPC 2D scan. The discharge was driven in 10 kPa synthetic air with sinusoidal voltage waveform at 6 kHz and a peak-to-peak amplitude of 2.4 kV. The gaseous gap was 0.9 mm, as depicted in figure 1. Spatial resolution was 0.1 x 0.1 mm² and thus a matrix of 12 x 12 was scanned point by point with TCSPC. From the data obtained one can carry out 2D emission distribution in the gap for a selected time of discharge development. Local emission diameter, streamer velocity at certain time or relative intensity of the radiation can also be evaluated.

![Figure 1](image.jpg)

**Figure 1.** The 2D development of the positive streamer luminosity in 10 kPa synthetic air recorded with sub-nanosecond resolution by a TCSPC device. One can resolve the creation of the streamer in the first sub-figure (on the left), its propagation in free space in the second and its impact on the cathode in the last sub-figure (on the right), where the spread of the surface discharges across the dielectric surface of the cathode is also shown.

In the case of experiments on laboratory TLE analogues at pressures below 2 kPa (the HV pulse-triggered streamers in amplitude-modulated AC-driven volume DBD with a discharge gap of 1 to 5 cm) we used a high-speed PMT to record streamer-induced emission through a set of band-pass filters. Under TLE-like conditions the temporal resolution of the PMT (Hamamatsu H10720) and spatial resolution of 1 mm seem to be enough for capturing streamer emission with sufficient precision and the use of complex TCSPC analysis can be avoided. In this case, the streamer and the PMT recording can be triggered externally by the same pulse/delay generator. Figure 2 illustrates the discharge luminosity scanned radially in the centre of the gap with spatial resolution of 1 mm. For each radial position a PMT signal waveform was recorded as an average of 1024 samples. Resulting 2D maps of projected streamer luminosities acquired with two selected band-pass filters are displayed in figure 2. Panel (a) in the figure shows the distribution of streamer filament luminosity at a wavelength corresponding to the (0,0) band of N₂-SPS and panel (b) shows luminosity at a wavelength corresponding to the (0,0) band of N₂⁺-FNS (corrected for spectral overlap with the SPS (2,5) band).
The use of TCSPC or pulse-triggered PMT methods means the emissions of selected spectral bands/lines of streamers can be evaluated. Typically, in nitrogen-oxygen mixtures the method of intensity ratio of first negative and second positive systems of molecular nitrogen can be used for electric field determination [4–6]. Similar measurements to those presented in figure 1 were performed for more challenging atmospheric pressure discharge conditions in the same set-up for emissions of second positive and first negative systems [7] and the 2D distribution development of the electric field was determined [8].

Figure 2. Radial scan of projected luminosity of triggered streamer generated in pure nitrogen at pressure of 1.33 kPa and in point-to-plane DBD gap of 1 cm. Panels (a) and (b) show SPS and FNS emission projections, respectively. The 2D maps were acquired from the middle of the gap.

3.3. **ICCD spectroscopy, imaging and microscopy**

ICCD-based techniques are used to provide spectral analysis of streamer luminosity when coupled with a dispersion spectrometer or to register streamer morphology when used as an imaging device, both with temporal resolution limited by the performance of the last-generation image intensifiers (typically 0.5 to 2 nanoseconds). Spectrometric analysis of streamer-induced emission can provide roto-vibronic distributions of diatomic species or relative emission intensities of various atomic/diatomic line/band systems [9]. Streamer morphology can be investigated through time-resolved (non)magnified ICCD images capturing 2D projection of the streamer luminosity. Because the excitation processes leading to the observed emissions strongly (exponentially) depend on the electric field, the streamer filament luminosity projected on and registered by the ICCD detector reflects characteristic dimensions of the streamer head.

A technique allowing acquisition of magnified images of surface micro-discharges was developed in [10]. Time-resolved microscopic images can be acquired synchronously either with the AC high-voltage voltage waveforms or micro-discharge current pulses. Figures 3, 4, 5 and 6 show typical examples of results obtained by ICCD microscopic images in a single coplanar surface dielectric barrier discharge (CSDBD) at atmospheric pressure. A detailed description of the reactor, CSDBD electrode geometry and related characteristics can be found elsewhere [10]. The images shown in
figure 3 were acquired with a large gate of ICCD intensifier (1s) synchronously with AC voltage and provide information on the spatial stability and influence of the oxygen addition on the micro-discharge morphology. It is worth pointing out that the ICCD images capture predominantly the SPS and FPS emissions of N2.

The ICCD images shown in figure 3 result from the accumulation of about 500 micro-discharges. The discharges seem to be diffused and no single streamer can be resolved because of the vertical spatial jitter of the micro-discharges and the increase of their diameter with the increasing content of oxygen [11]. This becomes clearer if we look at single streamer images such as those shown in figure 4. The images a) to d) were registered during positive (a,c) and negative (b,d) half-cycles in N2 (a,b) and N2-5%O2 (c,d). If we compare pictures taken in pure nitrogen with those taken in nitrogen-oxygen mixture the influence of oxygen on the diameter of the single streamer micro-discharge is evident.

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Figure 3. ICCD images registered with the exposure of 1 s at a fixed spatial resolution of 2.7 μm/pixel. The images show variations of the streamer radius together with spatial jitter for the case of non-triggered single surface streamer discharge in N2-20%O2 (a), N2-10%O2 (b) and N2-5%O2 (c) at atmospheric pressure.

Figure 4. ICCD microscopic images of surface CSDBD streamer in pure N2 (a-b) and in nitrogen with 5% admixture of oxygen (c-d), registered during positive (a-c) and negative (b-d) half-cycles of AC high-voltage waveforms ($u_{ac} = 23kV_{p-p}$) with exposure time of 50 μs. The grounded electrode and HV cathode are located on the left and right side, respectively.

Streamer propagation velocity, electric field in the streamer head and streamer diameter are closely related parameters, and knowledge of any two parameters could, in principle, be used to estimate the remaining one [12]. ICCD microscopic images can be used to determine the streamer head/channel diameter, the luminous streamer channel diameter and the propagation trajectory [10]. Defining the luminous minimum streamer diameter as the distance between the limits corresponding to the values of 10% and 90% of the integral of luminosity along the direction perpendicular to the streamer propagation axis [10], we estimated a minimum luminous CSDBD streamer diameter of 50 μm in pure N2 [10]. Adding oxygen to the gas mixture results in a larger streamer diameter. As an example, the typical luminous diameters obtained by applying former procedure in the case of a mixture of N2-5%O2 are reported in figure 5. The axial dependence (along the x-axis in figure 5) of the luminous diameter reveals, as in the previous cases, an asymmetric V-shape with a minimum diameter of about
70 µm located close to the temporary cathode, as for the streamer filaments produced both in N₂ and Ar [10].

Figure 5. ICCD microscopic image of single CSDBD event occurring in the N₂-5%O₂ mixture (\(u_{ac} = 23\text{kV}_{p-p}\)). The image (on the left) was used to estimate the axial evolution of the streamer luminous diameter (on the right). The (0,0) axis origin is placed in the centre of the gap. The grounded electrode and HV cathode are located on the left (\(x=-0.5\)) and right (\(x=+0.5\)) side, respectively.

In the case of cylindrically symmetrical discharge filaments, radial distributions of electronically excited species produced during surface streamer propagation can be obtained by applying the Abel inverse transform to projected luminosities of single streamer events [13], as shown in figure 5. Here, Abel inversion was performed for the projected profiles taken at \(x=-0.1\) mm coordinate of figure 5 following the procedure described in [13]. Projected as well as Abel-inverted profiles can be fitted by Gaussian functions with very similar half-widths (\(\sigma_1, \sigma_2\)), as shown in figure 6. In this case however the projected profile, in addition to the Abel-inverted one, can be well approximated only by the sum of two coaxial Gaussians with two different half-widths and weights.

Figure 6. Experimental projected and resultant Abel-inverted profiles (both symbols) together with the corresponding Gaussian fits (both solid lines) obtained from ICCD microscopic images (figure 5 of N₂-5%O₂) of a typical single CSDBD event at \(x=-0.1\)mm (corresponding to the axial position of the minimum diameter of the streamer filament).
An equivalent procedure was performed in the case of projected profiles shown in figure 2. Abel inversion [14] was performed for the projected profiles taken from figure 2 at time of 2 ns. In this case we inverted separately two different projected profiles corresponding to the FNS and SPS emissions. The projected (a,c) and Abel-inverted (b,d) profiles are shown in figure 7 together with the FNS/SPS ratio (e) of Abel-inverted profiles. Because ratio FNS/SPS ratio depends on the reduced electric field, the profile shown in panel (e) reflects radial evolution of electric field along the streamer radius in the middle of the gap at given time.

Figure 7. Experimental projected (a, c) and resultant Abel-inverted profiles (b, d) together with the corresponding Gaussian fits (both solid lines) obtained from radial scan of projected FNS and SPS luminosity of triggered streamer. Panels (a, c) show SPS while (b, d) FNS profiles. Panel (e) shows FNS/SPS ratio of inverted profiles. Abel inversion was performed for the projected profiles taken at position of maximum FNS intensity at conditions displayed in figure 2 (pure nitrogen at pressure of 1.33 kPa and in point-to-plane DBD gap of 1 cm).

3.4. LIF techniques and $N_2(A^3\Sigma_u^+)$ detection

The determination of absolute densities of transient species is quite a challenging task, as can be demonstrated in the case of $N_2(A^3\Sigma_u^+)$ metastables. The crucial role of these long-living species lies in the fact that they are important energy carriers capable of driving many energy transfers or chemical processes. In relation to streamer discharges probably the most versatile approach to detecting non-radiative species is laser-induced fluorescence (LIF). The detection of $N_2(A^3\Sigma_u^+, v=0-3)$ species may be based either on conventional one-photon LIF[15,16] or on two-photon optical-optical double-resonance (OODR) LIF techniques [17–19].

Recently, we investigated the temporal evolution of $N_2(A^3\Sigma_u^+, v=0-3)$ metastable species produced by a single triggered nitrogen streamer employing LIF and emission diagnostics simultaneously, and provided experimental evidence that maximum density of $N_2(A^3\Sigma_u^+, v=0-3)$ metastable species occurs on a microsecond timescale during the streamer channel decay [20]. It is worth pointing out that the single photon LIF technique can be successfully applied only in the case of precise timing of micro-discharge events, i.e. in the case of nanosecond-triggered discharges. In random events, a more sensitive technique might be applied, such as OODR-LIF [19] at the expense of a more complicated optical layout.
As an example, we show results obtained by applying the single photon LIF technique described in detail in [20] to monitor odd vibrational levels under discharge conditions illustrated in figure 2 (pure N₂, at 1.33kPa). Figure 8 shows the temporal behaviour of relative populations of individual N₂(A³Σ_u^+, v = 1, 3, 5, 7) vibronic levels normalized to their maximum. All four curves clearly show the formation of broad maxima shifted with respect to the streamer onset (t=0) with individual delays on a microsecond timescale and decreasing with the vibrational quantum number (about 50, 14, 2.3 and 1.4 µs for v = 1, 3, 5 and 7, respectively).

![Figure 8. Temporal evolution N₂(A³Σ_u^+, v) vibrational level for odd vibrational numbers produced by triggered streamer discharge in pure nitrogen at a pressure of 1.33 kPa and in point-to-plane DBD gap of 1 cm. The N₂(A³Σ_u^+, v) evolution is acquired from the middle of the gap.](image)

For proper interpretation of the experimental findings we need to understand the behaviour of metastable and other electronically excited N₂ species. Under streamer conditions, the discharge onset and propagation are an extremely fast event, which takes place on a (sub-)nanosecond timescale. As reported in [20], direct electron impact of the streamer head electrons establishes initial populations of individual vibrational levels which are redistributed because of radiative relaxation and heavy particle collisions on a much longer timescale (from units to hundreds of microseconds). It is well known that the N₂(A³Σ_u^+, v) metastable state serves as a terminal state for all N₂ triplet manifolds thanks to efficient intersystem collisional transfer (ICT) and redistribution within the N₂(A³Σ_u^+) state vibrational manifold (controlled by a specific process of vibrational relaxation).

The interpretation of similar experimental findings through a simplified numerical model implies that an important part of the metastable species becomes lost during the relaxation processes and estimation of N₂(A³Σ_u^+) state populations based on the LIF measurements of low vibrational levels could lead to significant undervaluation of the total metastable state population created during the streamer propagation phase [20].

4. Discussion and conclusions
To study the evolution of a developing streamer, optical diagnostics have to be performed with significantly enhanced spatio-temporal resolution. As regards radiative states, several approaches based on the 2-D projections of luminosities of cylindrically symmetrical streamers can be used to investigate spectrometric characteristics, the morphology of the streamer channel or radial distributions of electronically excited species within the streamer channel.
As regards non-radiative states, such as important long-living $\text{N}_2(A^3\Sigma_u^+, v)$ metastables, LIF-based techniques need to be applied to investigate the evolution of metastables on microsecond or even millisecond timescales. High vibrational levels of $\text{N}_2(A^3\Sigma_u^+, v)$ metastables under streamer conditions need to be studied in order to determine the total population of the $\text{N}_2(A^3\Sigma_u^+, v)$ manifold. However, the use of the discussed techniques under non-standard conditions imposed by micrometre spatial and picosecond timescales of streamers is always critical. Therefore, all the restrictions and limitations of the proposed techniques (timing and jitter issues) should be very carefully examined.

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