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High-Resolution Spectroscopic and Electrical Diagnostics of Barrier Discharges

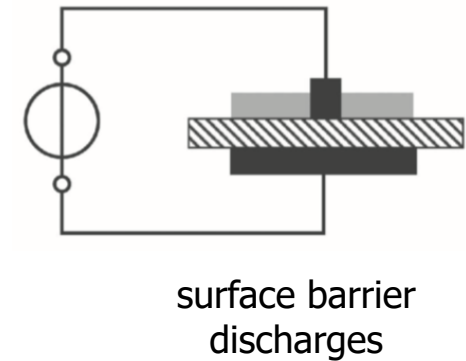
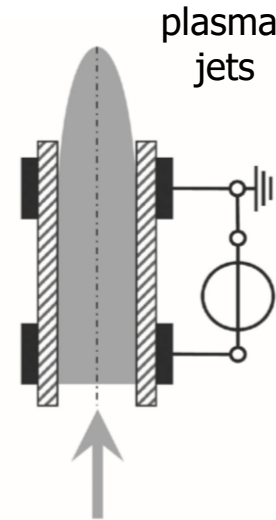
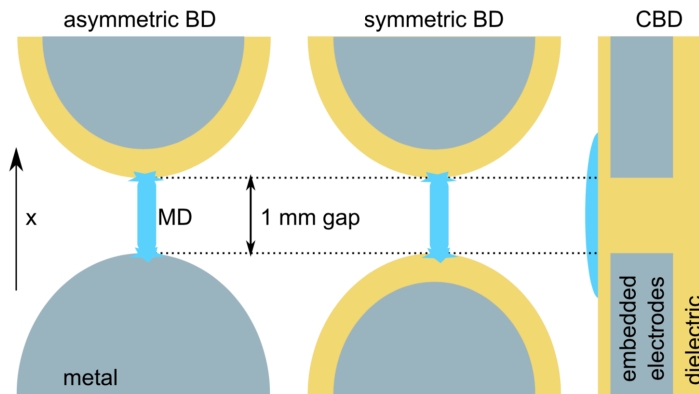
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- ❖ Barrier discharge and the streamer mechanism introduction
- ❖ The necessary resolution to catch the streamer discharge phenomenon at its characteristic scales in air at atmospheric pressure – **the instantaneous electric field quantification**
- ❖ Recent **advances in electrical measurements** on barrier discharges
- ❖ Application on surface barrier discharge in contact with water

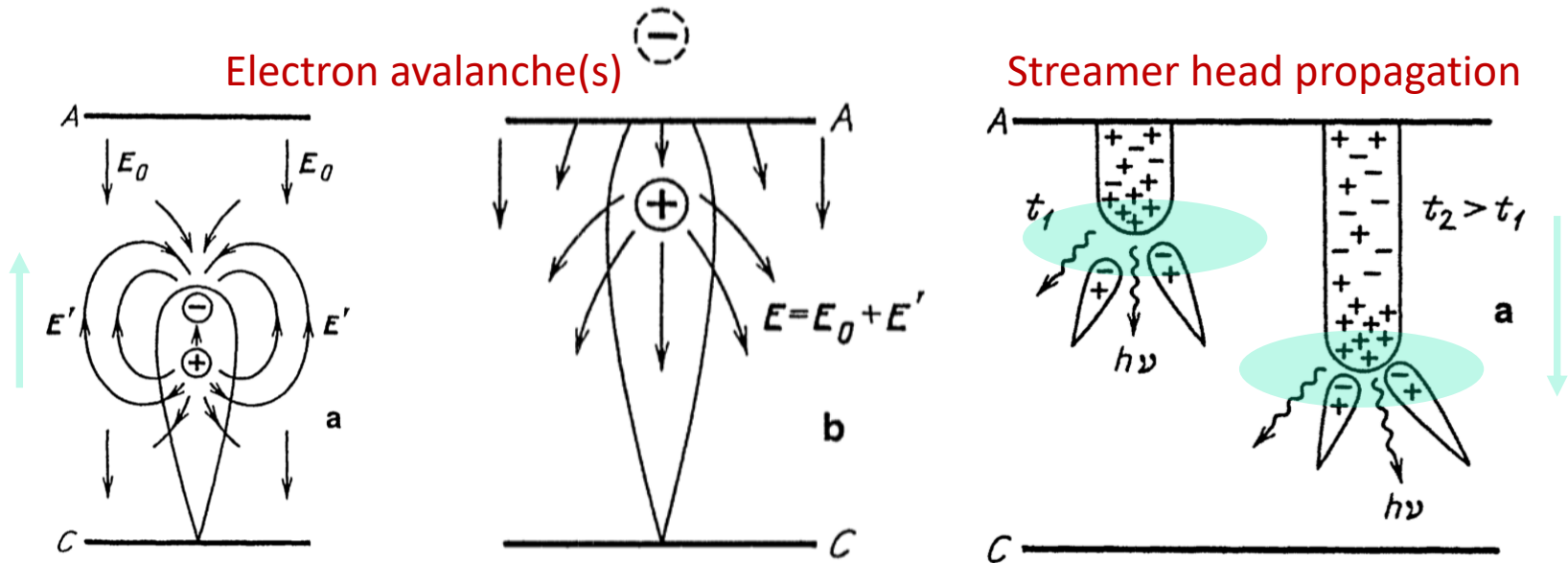
Barrier discharge



- Presence of insulating barrier between the electrodes
- Typically AC or pulsed applied voltage waveform
- Different geometries for different uses: volume barrier discharges for ozone production, plasma jets for use in medicine, water treatment, coplanar type for surface treatment...

Kogelschatz 2002 PSST
Wagner 2003 Vacuum
Černák et al. 2011 PPCF
Brandenburg 2017 PSST

Streamer discharge



- Streamer is contracted **fast moving ionizing wave**.
- Streamer is characterized by a **self-generated electric field** enhancement at the head of the growing discharge channel, leaving a filamentary plasma behind.
- Usually it results **from the space charge left by electron avalanches**.
- Streamers in barrier or corona streamer microdischarges in air at atmospheric pressure are, however, challenging the standard technique resolutions – **duration only few units of ns**.

Raizer Yu 1997 Gas discharge physics
 Hodges et al. 1985 Phys. Rev. A
 Ebert et al. 2006 PSST
 Marode et al. 2009 PPCF

Resolution in time and space and what is possible today?

The nanosecond resolution of ICCD devices is a standard and a broad variety of spectroscopic methods for basic plasma parameter determination is at disposal.

IOP Publishing

J. Phys. D: Appl. Phys. 47 (2014) 485201 (13pp)

Journal of Physics D: Applied Physics

doi:10.1088/0022-3727/47/48/485201

An electric field in nanosecond surface dielectric barrier discharge at different polarities of the high voltage pulse: spectroscopy measurements and numerical modeling

S A Stepanyan¹, V R Soloviev² and S M Starikovskaia¹

IOP Publishing

Plasma Sources Sci. Technol. 24 (2015) 034001 (18pp)

Plasma Sources Science and Technology

doi:10.1088/0963-0252/24/3/034001

Electron density measurement in atmospheric pressure plasma jets: Stark broadening of hydrogenated and non-hydrogenated lines

A Yu Nikiforov^{1,2}, Ch Leys¹, M A Gonzalez¹ and J L Walsh¹

IOP Publishing

Plasma Sources Sci. Technol. 23 (2014) 015011 (12pp)

Plasma Sources Science and Technology

doi:10.1088/0963-0252/23/1/015011

Optical spectroscopy diagnostics of discharges at atmospheric pressure

Giorgio Dilecce

IOP Publishing

J. Phys. D: Appl. Phys. 46 (2013) 464001 (28pp)

JOURNAL OF PHYSICS D: APPLIED PHYSICS

doi:10.1088/0022-3727/46/46/464001

Atmospheric pressure discharge filaments and microplasmas: physics, chemistry and diagnostics

Peter Bruggeman^{1,2} and Ronny Brandenburg^{1,3}

IOP PUBLISHING

J. Phys. D: Appl. Phys. 45 (2012) 295201 (11pp)

JOURNAL OF PHYSICS D: APPLIED PHYSICS

doi:10.1088/0022-3727/45/29/295201

Measurement of the temporal evolution of electron density in a nanosecond pulsed argon microplasma: using both Stark broadening and an OES line-ratio method

Xi-Ming Zhu¹, James L Walsh², Wen-Cong Chen¹ and Yi-Kang Pu¹

IOP Publishing

J. Phys. D: Appl. Phys. 47 (2014) 463001 (31pp)

Journal of Physics D: Applied Physics

doi:10.1088/0022-3727/47/46/463001

Topical Review

Optical diagnostics of streamer discharges in atmospheric gases

M Šimek

APPLIED PHYSICS LETTERS 99, 161502 (2011)

Spectroscopic measurement of electric field in atmospheric-pressure plasma jet operating in bullet mode

Goran B. Sretenović,^{ab} Ivan B. Krstić, Vesna V. Kovačević, Bratislav M. Obradović, and Milorad M. Kuraica

IOP PUBLISHING

Plasma Sources Sci. Technol. 12 (2003) 125–138

PLASMA SOURCES SCIENCE AND TECHNOLOGY

PII: S0963-0252(03)58383-9

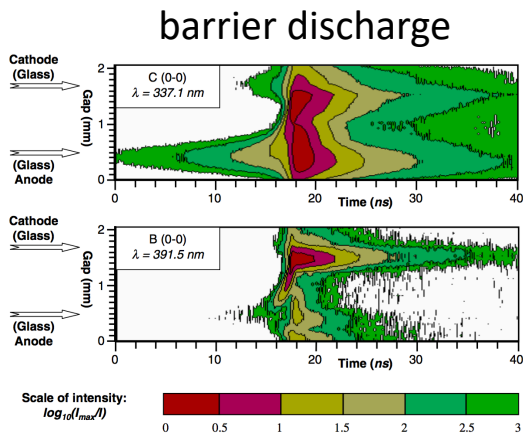
Optical diagnostics of atmospheric pressure air plasmas

C O Laux^{1,2}, T G Spence³, C H Kruger and R N Zare⁴

Streamer discharges and picosecond signal recording

The barrier or corona streamer discharges in air are, however, challenging the standard technique resolutions – duration only few units of ns.

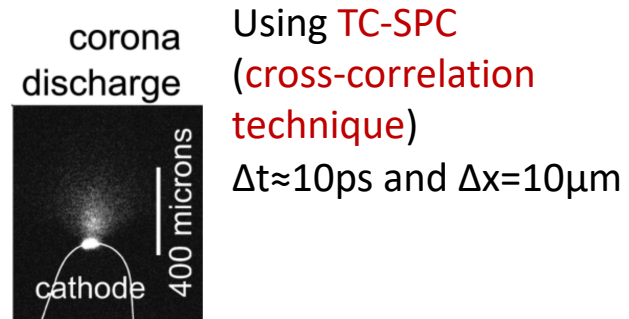
→ need for resolution in picosecond timescales!



Using **TC-SPC**
(**cross-correlation technique**)

$\Delta t < 0.1 \text{ ns}$
 $\Delta x = 100 \mu\text{m}$

Kozlov et al. 2001
J. Phys. D: Appl. Phys.

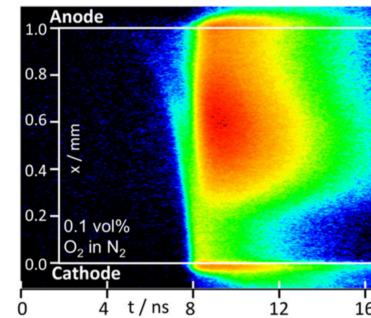


Using **TC-SPC**
(**cross-correlation technique**)

$\Delta t \approx 10 \text{ ps}$ and $\Delta x = 10 \mu\text{m}$

Hoder et al. 2012 Phys. Rev. E

pulsed barrier discharge



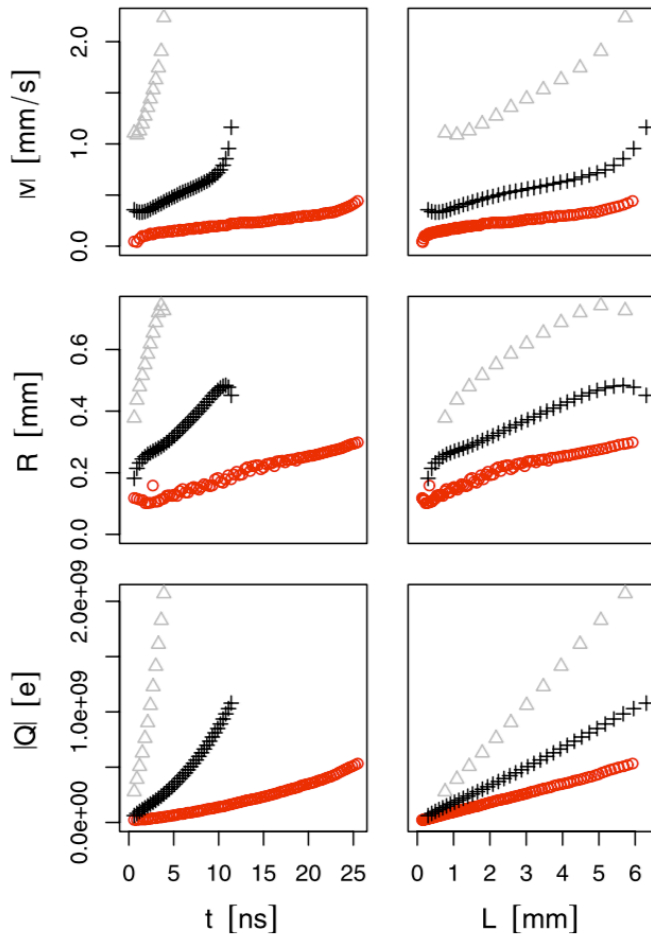
Using **streak camera coupled with far-field microscope**

$\Delta t \approx 50 \text{ ps}$ and $\Delta x = 10 \mu\text{m}$

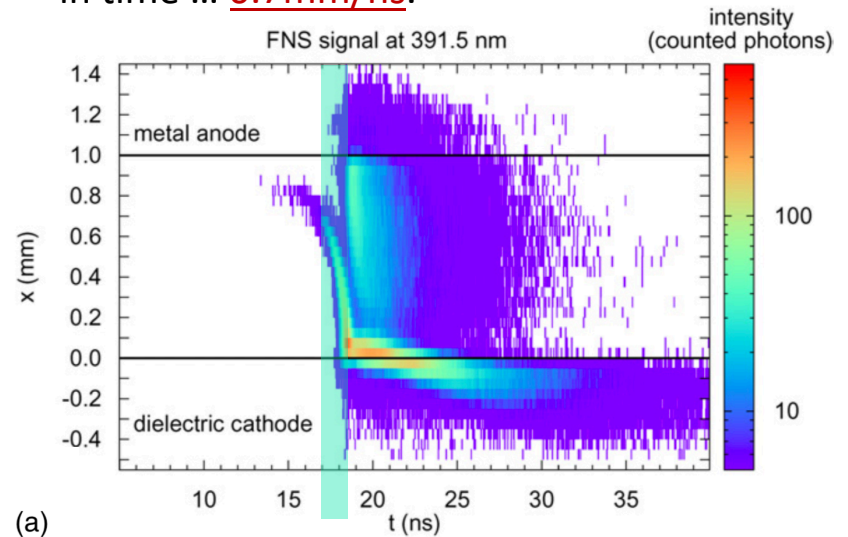
Höft et al. 2013 J. Phys. D: Appl. Phys.

Streamer discharge in atmospheric pressure air

Different voltages 21kV, 14kV, 10.5kV



Based on modelling - various phenomena takes place during the few nanoseconds of the streamer lifespan: streamer acceleration, its head is expanding, the amount of the net charge in the head is increasing, ...
 ... if you are lucky however, you can catch experimentally at least its 1D development in time ... 0.7mm/ns.



Luque et al. 2008 J.Phys.D:Appl.Phys.
 Hoder et al. 2010 J.Phys.D:Appl.Phys.

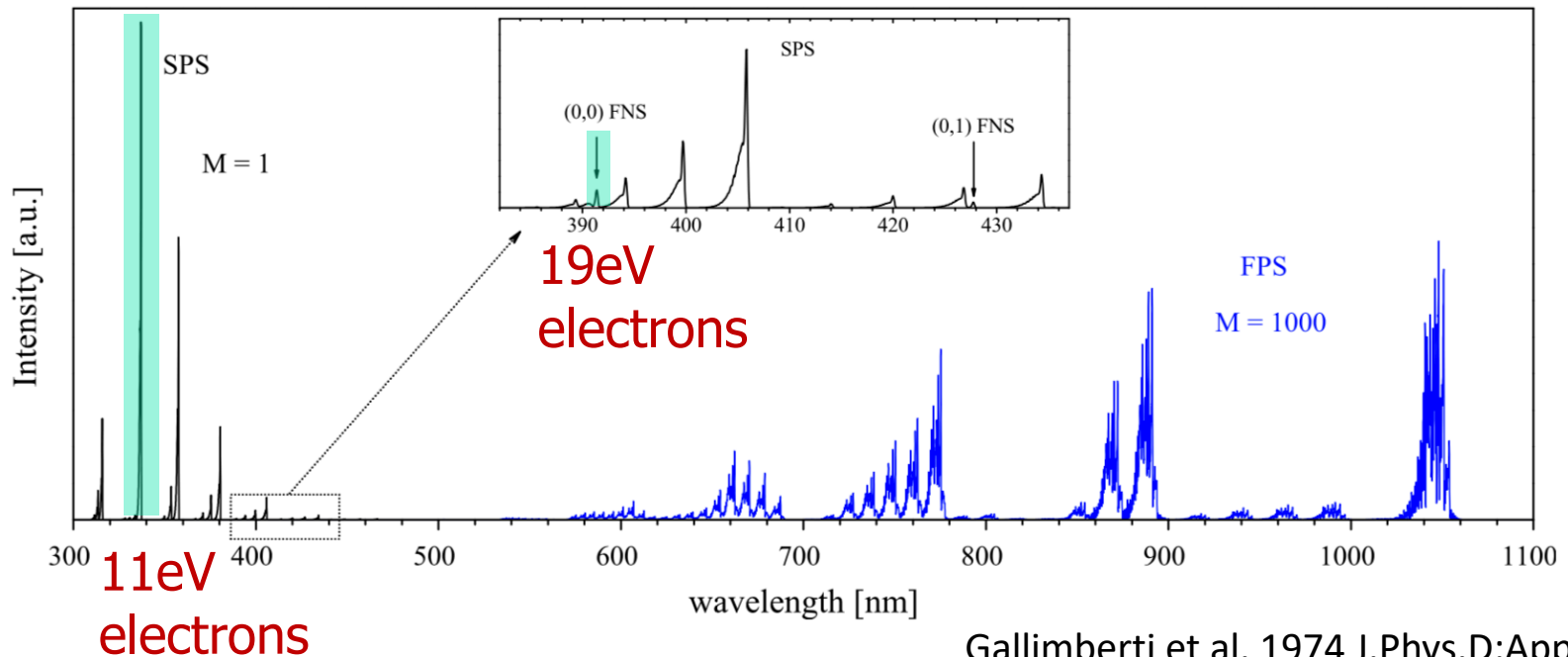
Optical emission spectroscopy on streamers in air

- towards the electric field

- in barrier discharges, coronas, lightning or transient luminous events (Red Sprites, Blue Jets) in atmospheric air, the ratio of intensities of first negative and second positive systems of molecular nitrogen is strongly dependent on E/N

$$\left(\frac{dI_{FNS}/dt + I_{FNS}/\tau_{eff}^{FNS}}{dI_{SPS}/dt + I_{SPS}/\tau_{eff}^{SPS}} \right) \frac{\tau_{eff}^{FNS}}{\tau_{eff}^{SPS}} = R_{FNS/SPS}(E/N)$$

Instantaneous development from non-steady-state kinetic model.



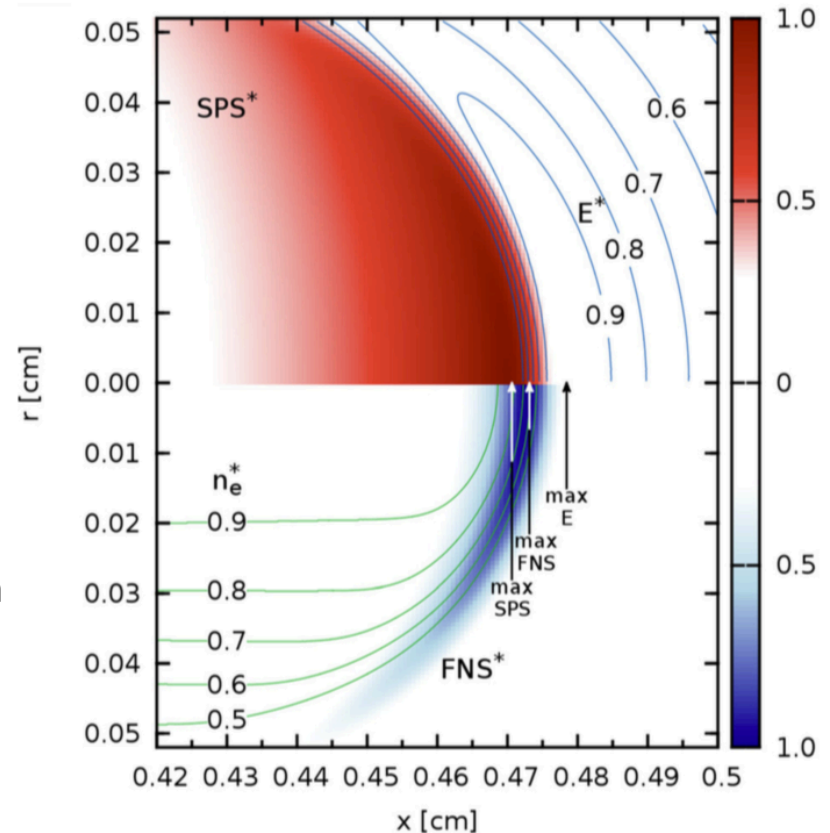
Gallimberti et al. 1974 J.Phys.D:Appl.Phys.
 Kozlov et al. 2001 J.Phys.D:Appl.Phys.
 Hoder, Bonaventura et al. 2015 J.Appl.Phys.

Optical emission spectroscopy on streamers in air - detailed know-how

- in barrier discharges, coronas, lightning or transient luminous events (Red Sprites, Blue Jets) **in atmospheric air, the ratio of intensities of first negative and second positive systems of molecular nitrogen is strongly dependent on E/N**

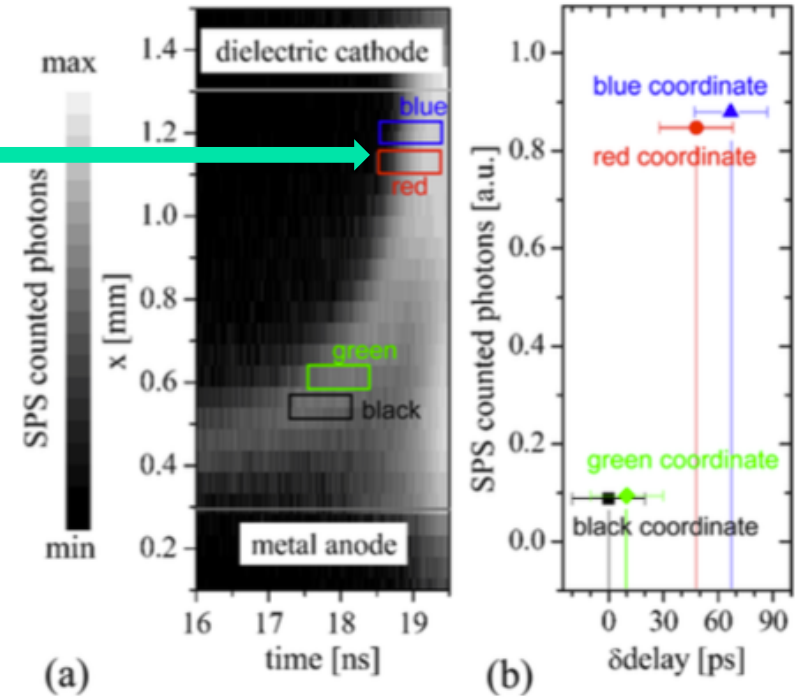
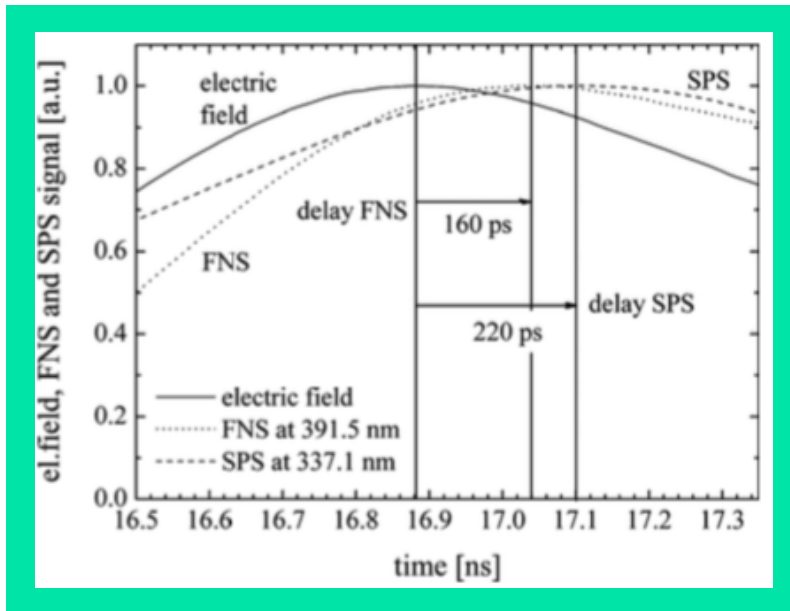
$$\left(\frac{dI_{\text{FNS}}/dt + I_{\text{FNS}}/\tau_{\text{eff}}^{\text{FNS}}}{dI_{\text{SPS}}/dt + I_{\text{SPS}}/\tau_{\text{eff}}^{\text{SPS}}} \right) \frac{\tau_{\text{eff}}^{\text{FNS}}}{\tau_{\text{eff}}^{\text{SPS}}} = R_{\text{FNS}/\text{SPS}}(E/N)$$

- ❖ relaxation of electron energy distribution function
- ❖ proper selection of quenching constants
- ❖ rotational temperature dependence of FNS/SPS
- ❖ need for tens of microns and picoseconds spatiotemporal resolution
- ❖ optimized kinetic model etc.



Hoder, Loffhagen et al. 2016 Plasma Sources Sci. Technol.
 Hoder, Šimek et al. 2016 Plasma Sources Sci. Technol.
 Hoder, Bonaventura, Bourdon et al. 2015 J.Appl.Phys.

Experimentally studied microphysics of the streamer in barrier discharge – basis for E/N determination



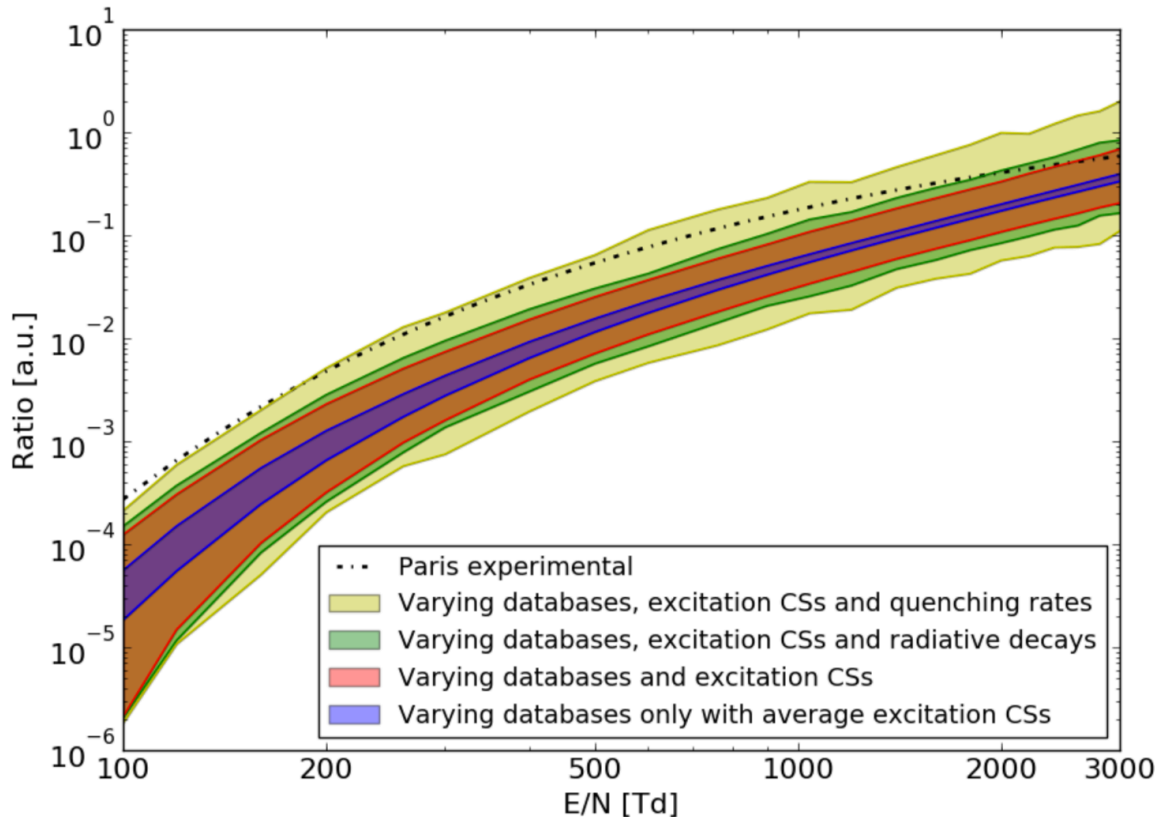
- Experimentally obtained mutual delay between the peak of spectrally resolved emissions and the electric field
- Dilatation/increase of this delay give us the measure of the streamer expansion – confirmed by 1D and 2D streamer models

Naidis 2009 Phys. Rev. E

Hoder, Bonaventura et al. 2015 J.Appl.Phys.

Optical emission spectroscopy on streamers in air - limitations and challenges

$$\left(\frac{dI_{\text{FNS}}/dt + I_{\text{FNS}}/\tau_{\text{eff}}^{\text{FNS}}}{dI_{\text{SPS}}/dt + I_{\text{SPS}}/\tau_{\text{eff}}^{\text{SPS}}} \right) \frac{\tau_{\text{eff}}^{\text{FNS}}}{\tau_{\text{eff}}^{\text{SPS}}} = R_{\text{FNS/SPS}}(E/N)$$



- **Uncertainty quantification**, localisation of **its main sources** and **uncertainty limitation** by using well selected cross-sections and life-times
- Usually used Paris's formula slightly underestimates the el. field value

Obrusník, Bílek et al. 2018 submitted
Paris et al. 2005 J.Phys.D:Appl.Phys.

Experimentally studied microphysics of the streamer in barrier discharge – basis for E/N determination

Based on the previous knowledge from the picosecond spectroscopy we can:

- Locate the streamer head with high precision
- Determine its electric field waveform shape with high resolution and
- Quantify its amplitude with quantified uncertainty
(knowing also where the uncertainty comes from)

We also obtained reliable values for the peak electric field in the streamer heads for barrier discharges in different arrangements:

- Confirming the intervals of typical values given by fluid and hybrid models
- Around 500Td (approx. 120kV/cm) for volume streamers
- And around 1200Td (approx. 300kV/cm) for surface streamers

Important know-how for remote electric field determination in microscopic discharges (where accurate laser spectroscopy has insufficient absorption path) or as a fundamental knowledge for atmospheric electricity investigation.

Hoder, Bonaventura et al. 2016 PSST
 Hoder, Synek et al. 2017 PPCF
 Stepanyan et al. 2014 J.Phys.D:Appl.Phys.
 Obrusník, Bílek et al. 2018 submitted
 Babaeva et al. 2016 PSST
 Luque et al. 2008 J.Phys.D:Appl.Phys.

Experimental study of electric current of the barrier discharge - typical cases

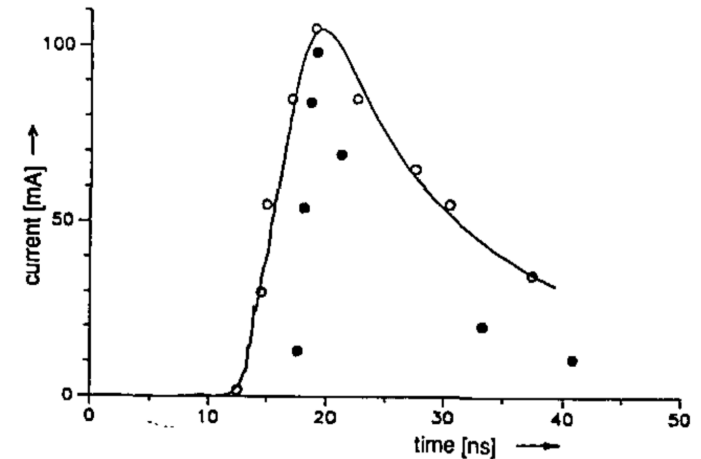
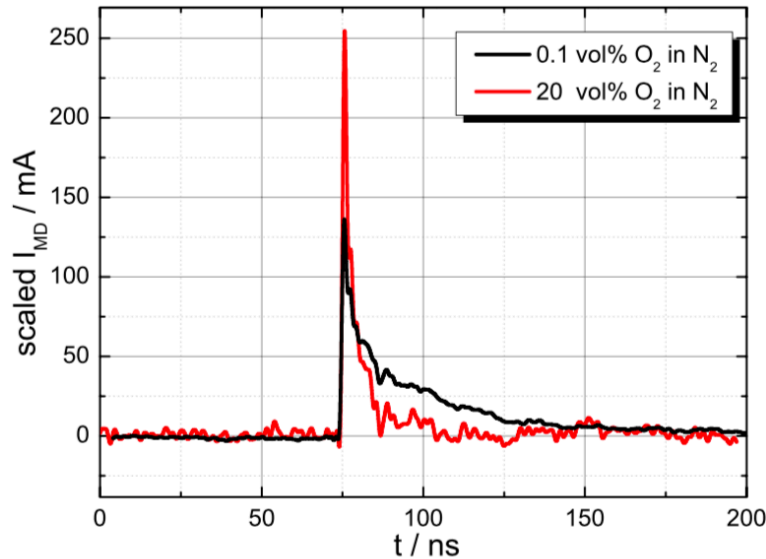
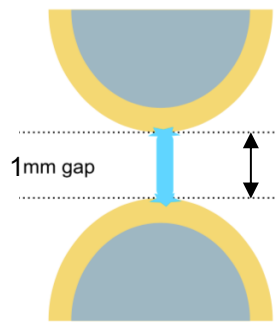
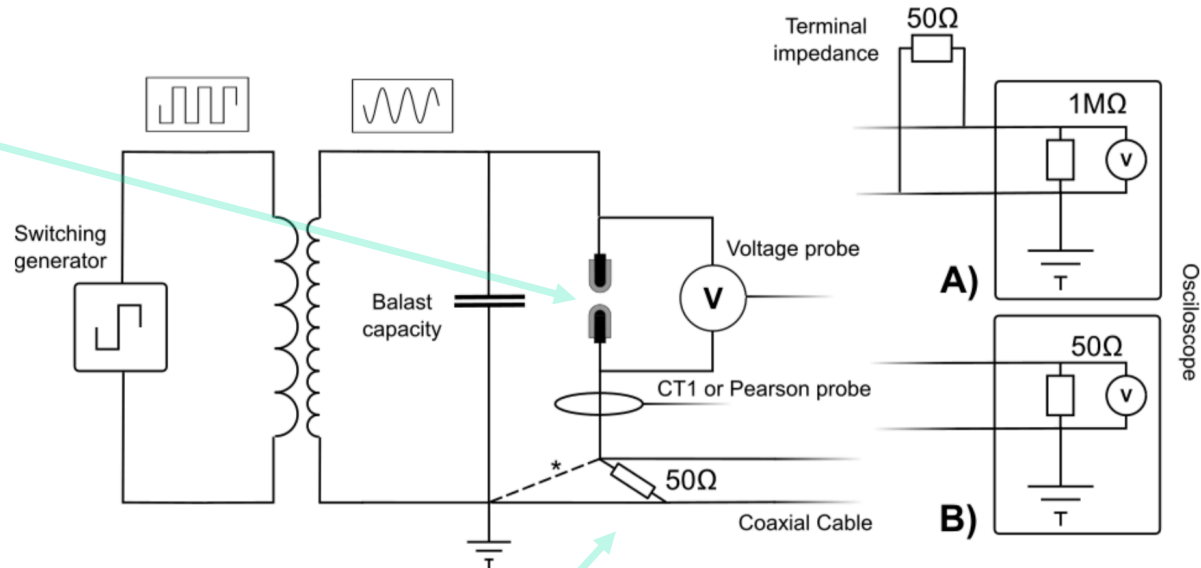
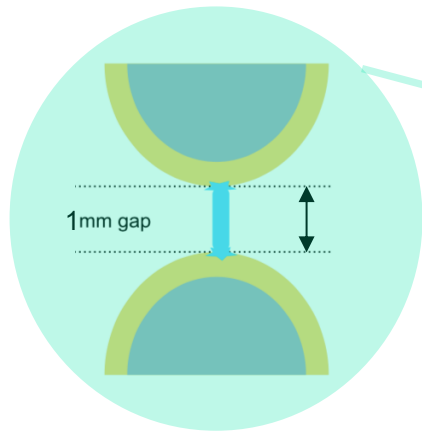


Figure 16. Comparison of calculated (full line) and measured (symbols) microdischarge currents. The measured values follow from similar boundary conditions (●: 1.2 bar instead of 1 bar [14], ○: 4 mm discharge gap [17]; the measured values are divided by 4).

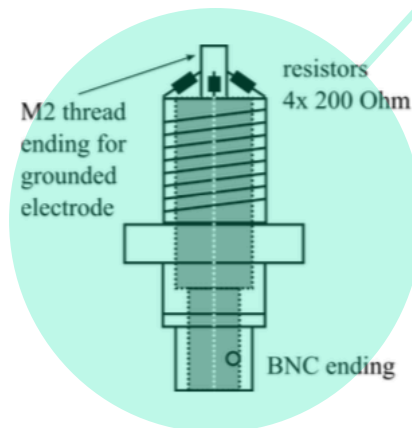
- Typical current pulse recorded by Rogowski-type current probes or on resistor shunts – **uncertainty in several milliamperes**
- The fast rise (few nanoseconds), sharp peak and the exponential decay is known – **the finer structure of the current pulse known from models is hidden in the noise**

Höft et al. 2013 J.Phys.D:Appl.Phys.
 Braun et al 1992 PSST

Electrical current measured by self-assembled current probe



a)



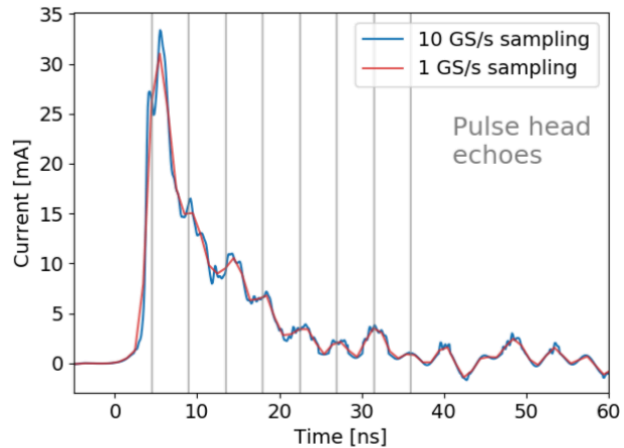
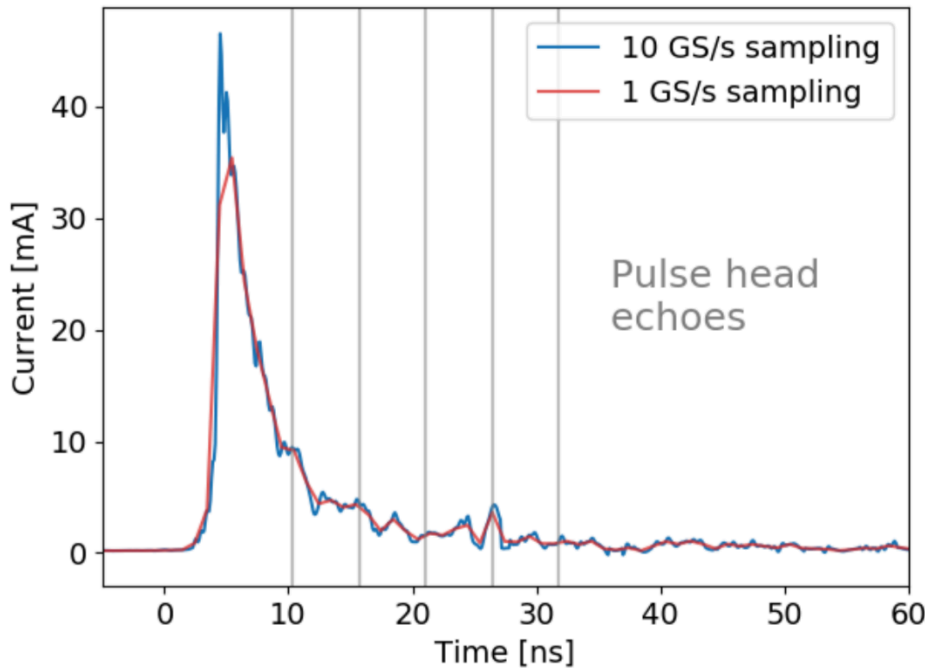
b)

- Resistors placed in coaxial arrangement
- Signal recorded through **coaxial cable**
- Measurement of the **voltage drop on total resistivity** of input 50Ohm and internal resistivity of the oscilloscope input :-O

Synek et al. 2018 submitted
Černák et al. 1993 J.Appl.Phys.

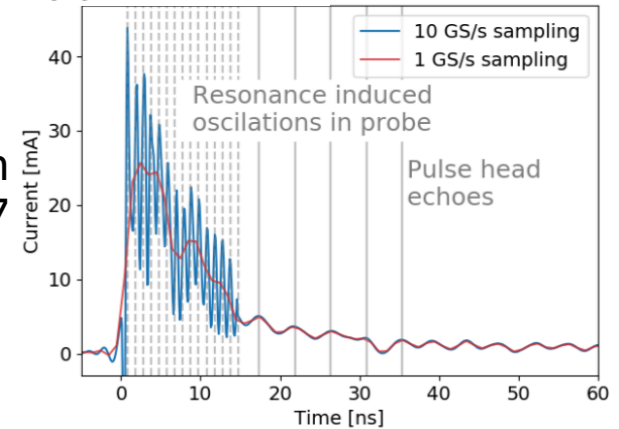
Electrical current measured by self-assembled current probe

self-assembled current probe



CT-1

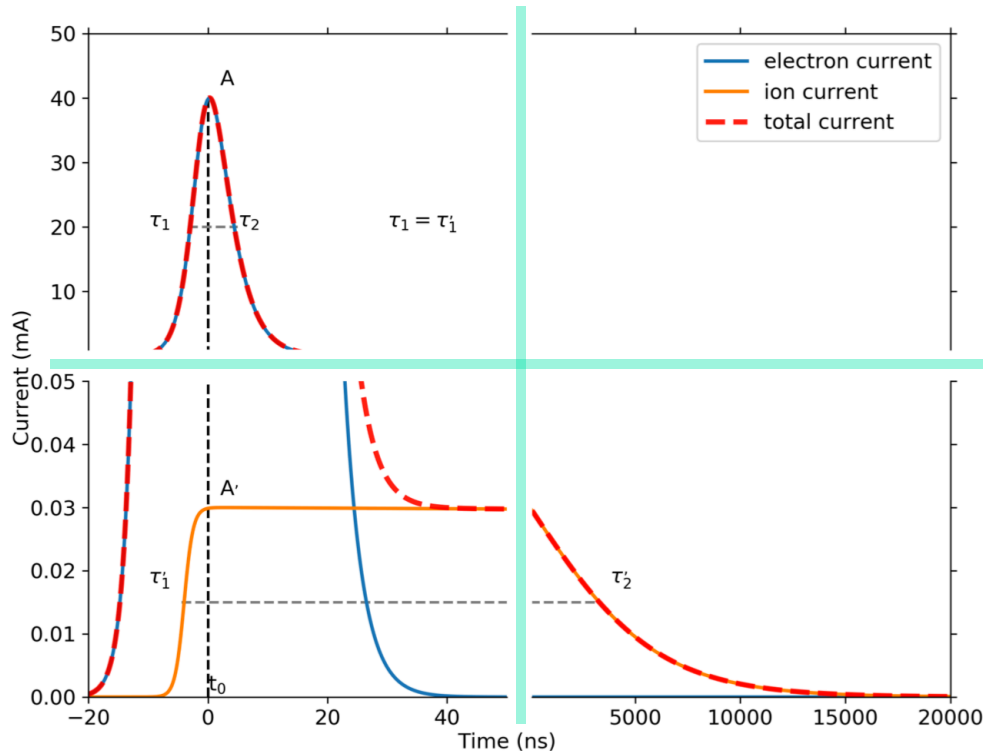
Pearson 2877



- Significantly **higher bandwidth of the probe** compared to Pearson 2877 or CT-1 probes, pulse FWHM decreases from 10ns for Pearson to 2.8ns for self-assembled probe
- **Increased signal-to-noise ratio**, sensitivity in several units of microamperes!

Synek et al. 2018 submitted

Estimated separation of electrical current for electrons and ions



$$f_{sigmoid}(t) = \frac{1}{1 + e^{-\frac{t_0-t}{\tau_1}}}$$

$$f_{exp.decay}(t) = \begin{cases} 1 & \text{for } t \leq t_0 \\ e^{-\frac{t_0-t}{\tau_2}} & \text{for } t > t_0 \end{cases}$$

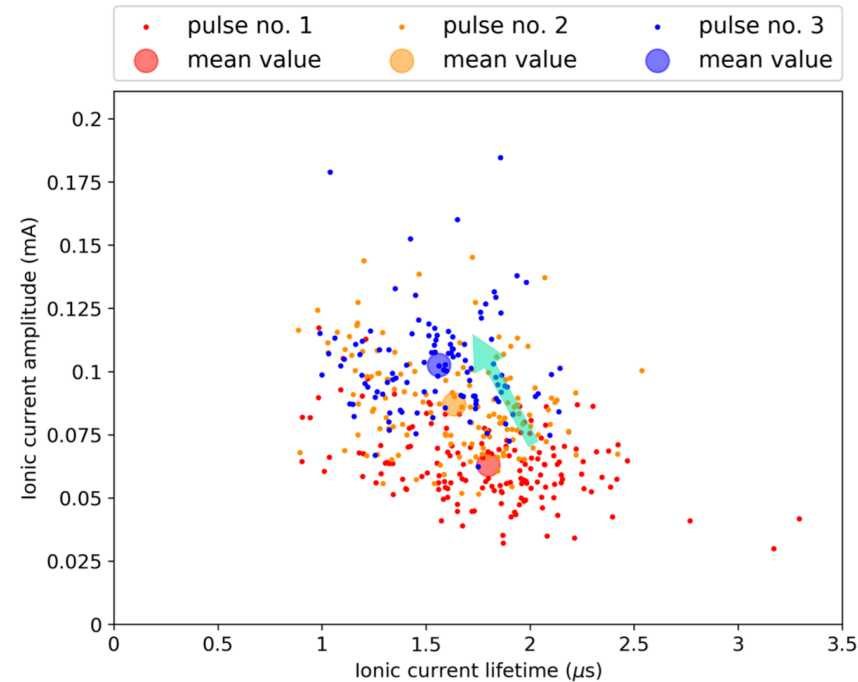
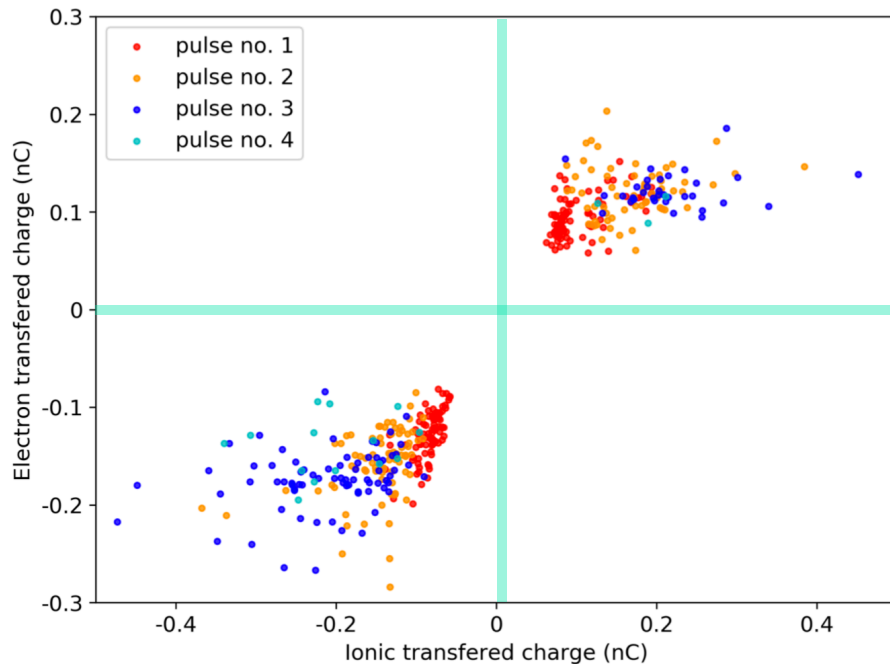
$$f_{profile}(t) = A \cdot f_{sigmoid}(t) \cdot f_{exp.decay}(t)$$

Each current pulse is a summation of two profiles, for electrons and for ions:

$$f(t)_{total} = f_{profile.el}(t) + f_{profile.ion}(t)$$

- **Electron current** is represented by intense short profile, while the **ionic current** is of low amplitude (tens of microamperes) and much longer decay (approx. 200times)
- Complication of the **unknown displacement current**, for its solution an appropriate computer model is necessary

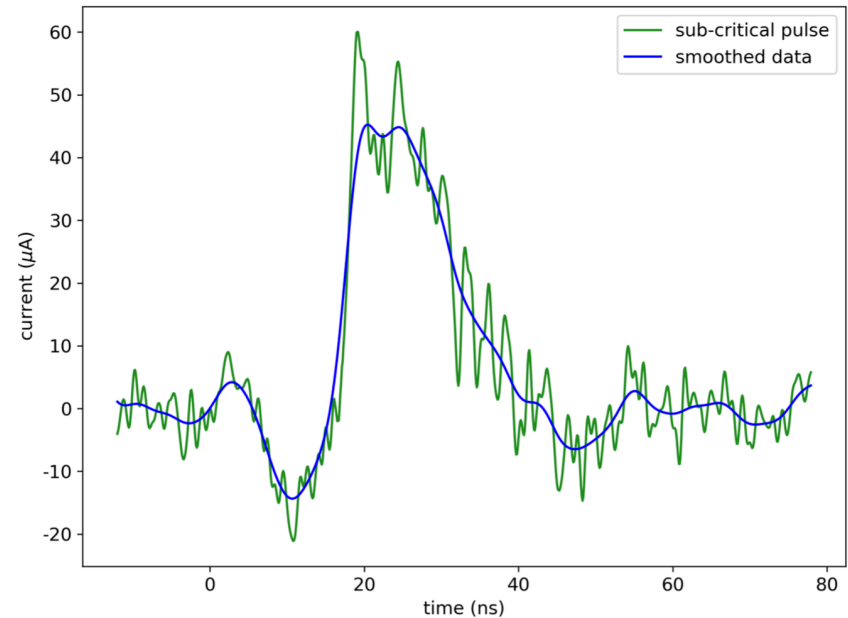
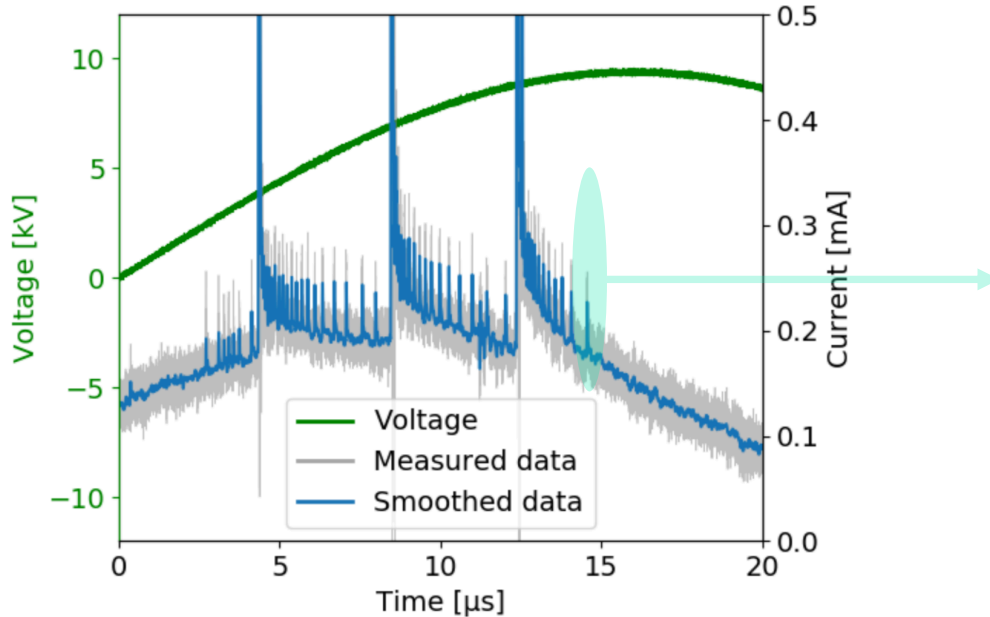
Statistical analysis of the electrical current for electrons and ions



- **Electron current** is comparable with the **ionic current** or several subsequent pulses with small variation – as expected from charge equilibrium
- The **amplitude is increasing and decay of ionic current shortening for subsequent current pulses** – increased conductivity due to the local heating or discharge mechanism change/modification

Synek et al. 2018 submitted

Evidence of repetitive micro-pulses, described as sub-critical pulses



- Amplitude of **few tens of microamperes**, transferred charge of **0.5pC**, i.e. approximately **10^7 electrons**, **sub-critical with respect to the Raether-Meek threshold** and in comparison to the microdischarge bridging the gas gap
- The amplitude remains almost stable, the **frequency is changing with the changing local electric field – hypothetically due to the amount of drifted ionic charge and/or the local electric field at the residual charge domains**

Synek et al. 2018 submitted

Results of electrical current measured by self-assembled current probe

Based on the enhanced electrical current measurements we can:

- Estimation of the separated electronic and ionic current components
- Determine the current with almost microampere sensitivity
- Quantify the transferred charge to sub-picocoulomb amounts (corresponding to sub-Raether-Meek amount of 10^7 electrons)

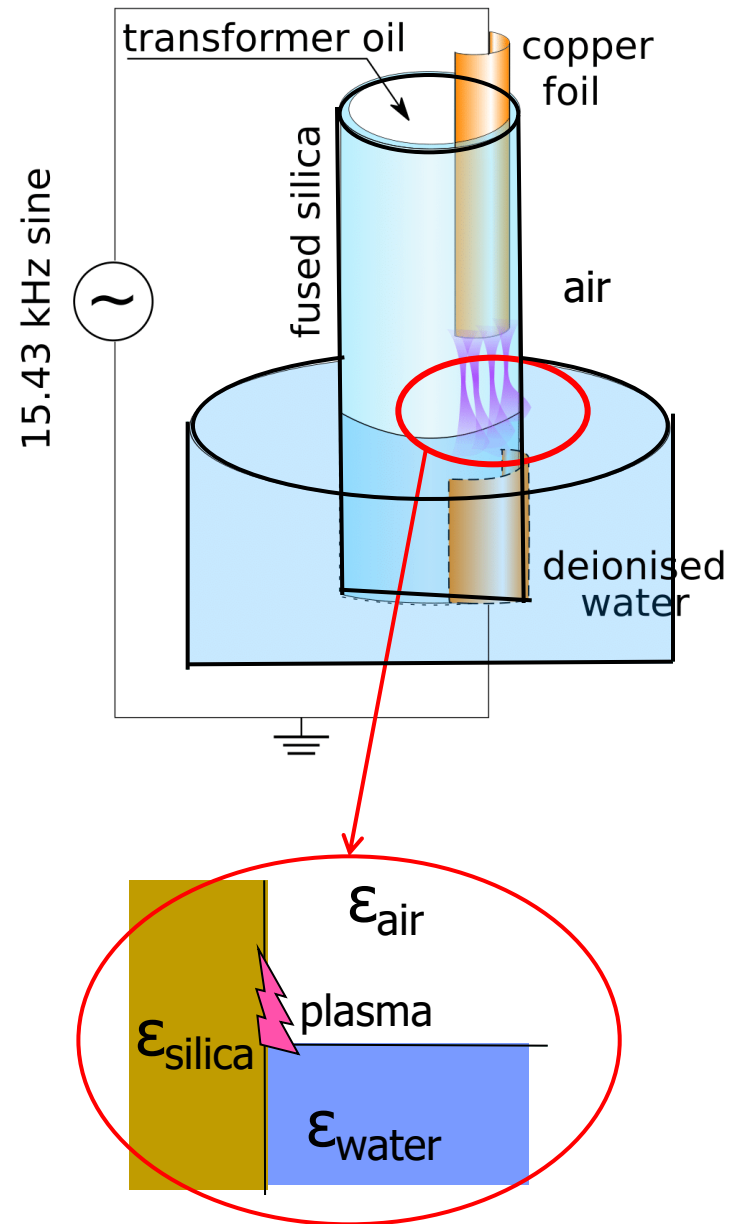
We also obtained new knowledge about the statistical behavior of the current pulses in volume barrier discharge:

- Evidence of new phenomena responsible for change in ionic current amplitude and decay
- Probably heating of the gas within one half-period or discharge mechanism variation/modification
- Detection of repetitive micro-pulses – hypothesis of discharging of residual surface charge micro-domains

Barrier discharge in air at water interface

- Both **electrodes** are not in contact with **plasma**.
- Plasma originates from **triple-junction** – line where the liquid (**de-ionized water**), solid (**fused silica cuvette**) and gas (**air with water vapor**) meet
- 13 kV peak-to-peak voltage
- 15.4 kHz sine frequency
- Chamber rinsed with 1 slm of air

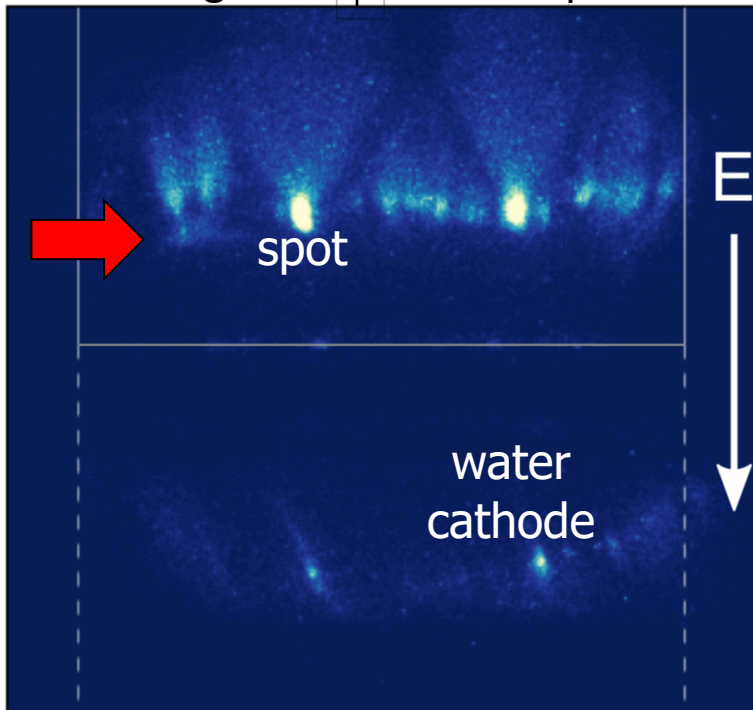
Voráč et al. 2017 J. Phys. D: Appl. Phys.
 Pavliňák et al. 2014 Appl. Phys. Lett.
 Galmiz et al. 2017 Plas. Proc. Polym.
 Galmiz et al. 2016 J. Phys. D: Appl. Phys.



ICCD imaging – discharge morphology

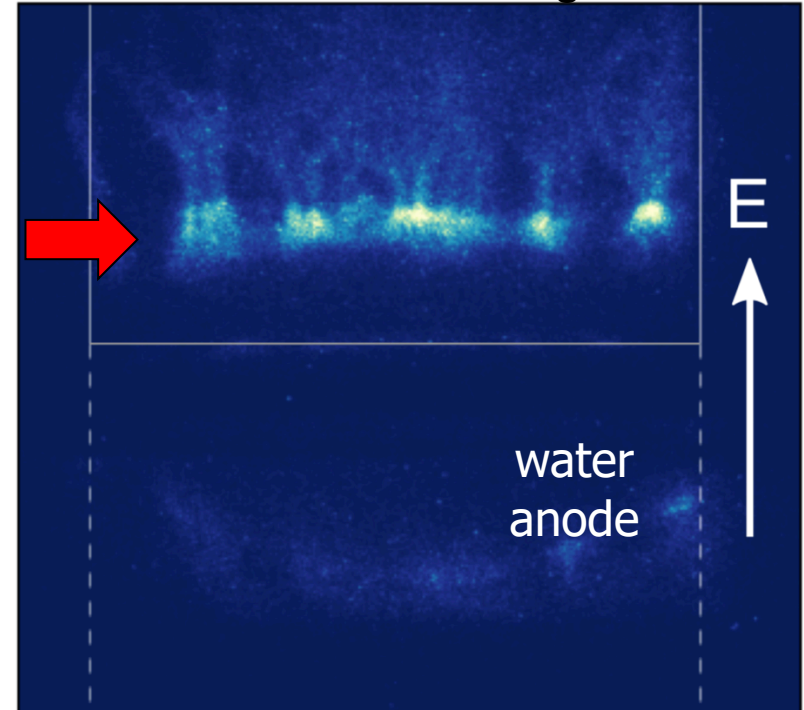
Negative streamer

discharges with cathode spots



Positive streamer

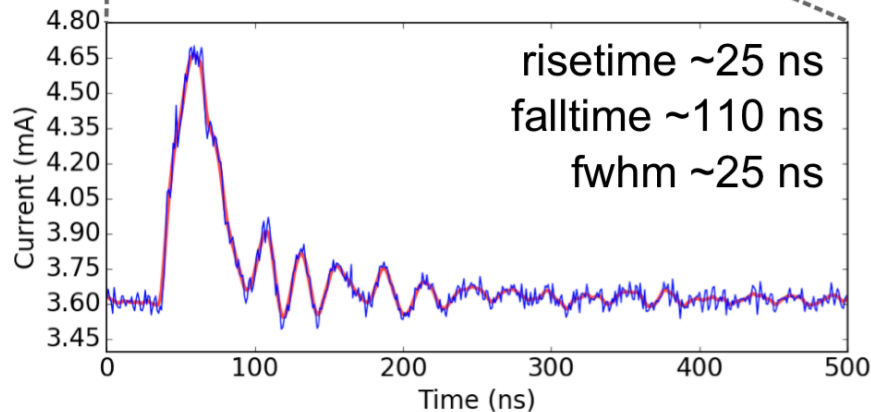
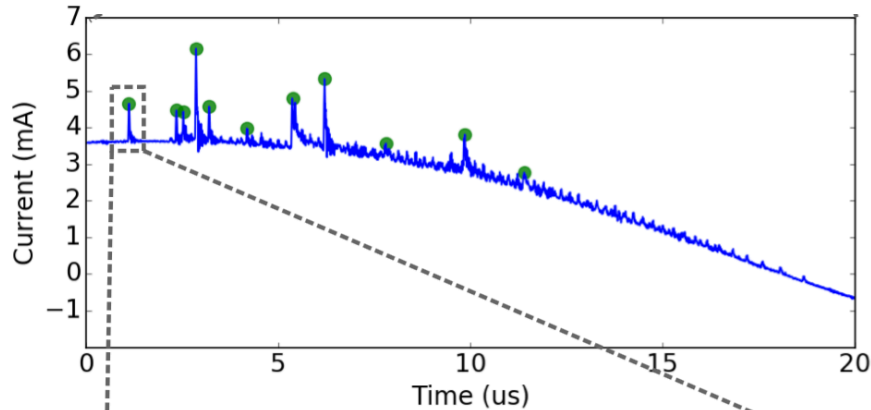
branched discharges



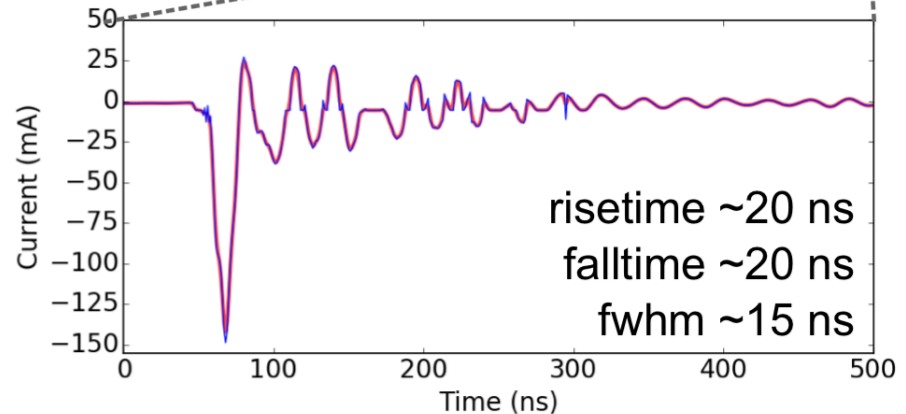
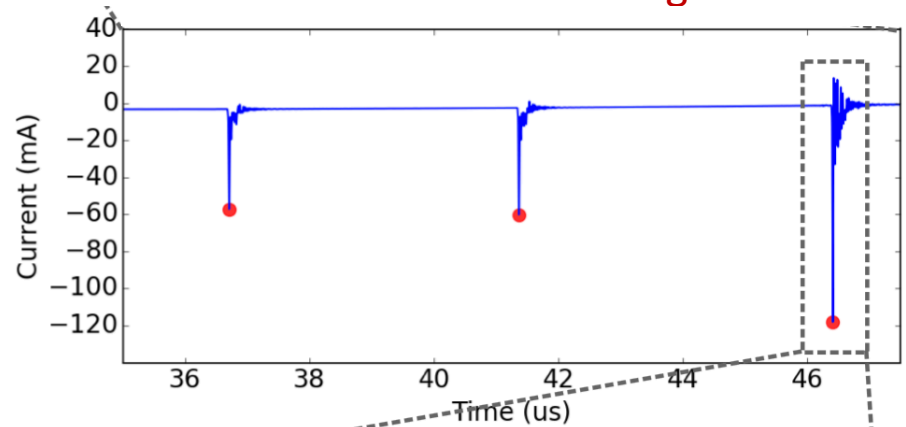
- water level is marked by horizontal grey line
- **triple-line marked by red line**, is raised due to capillary effect

Detailed comparison of current pulses

Negative streamer discharges



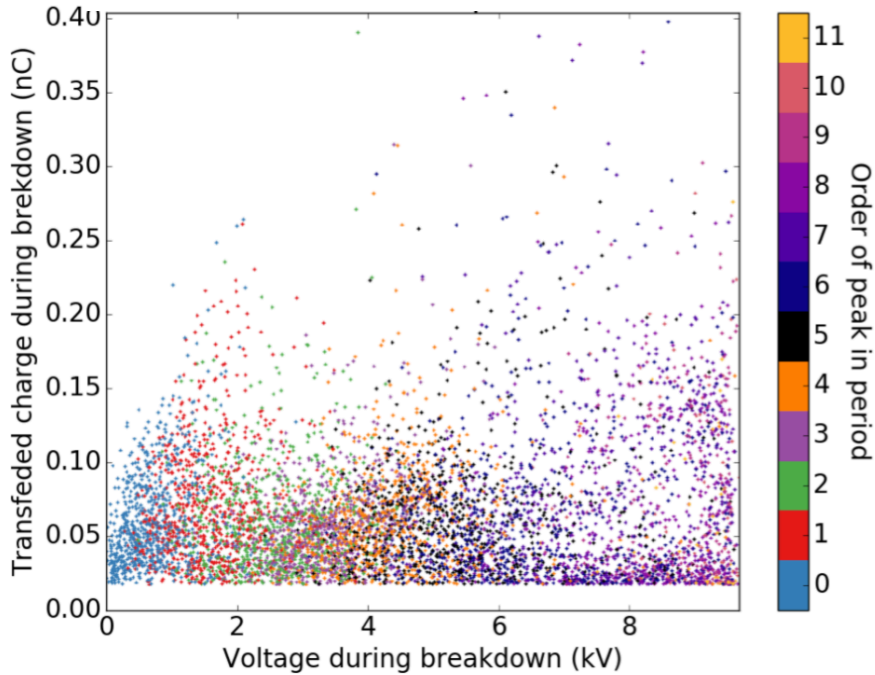
Positive streamer discharges



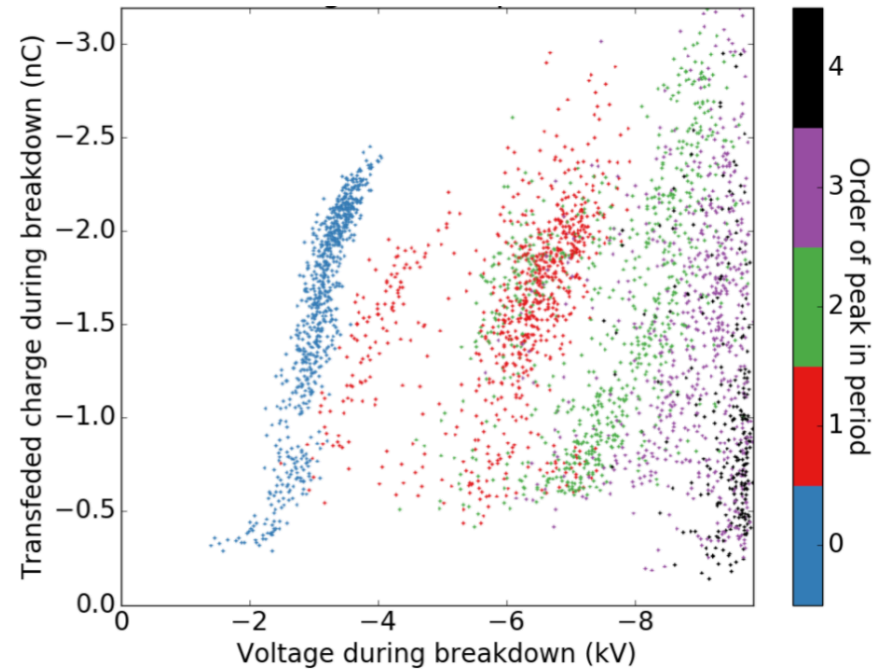
- Expected behavior from the surface barrier discharges with solid metallic electrodes
- Variations of small pulses for negative discharges and large distinct pulses for positive

Statistical comparison of current pulses

Negative streamer discharges



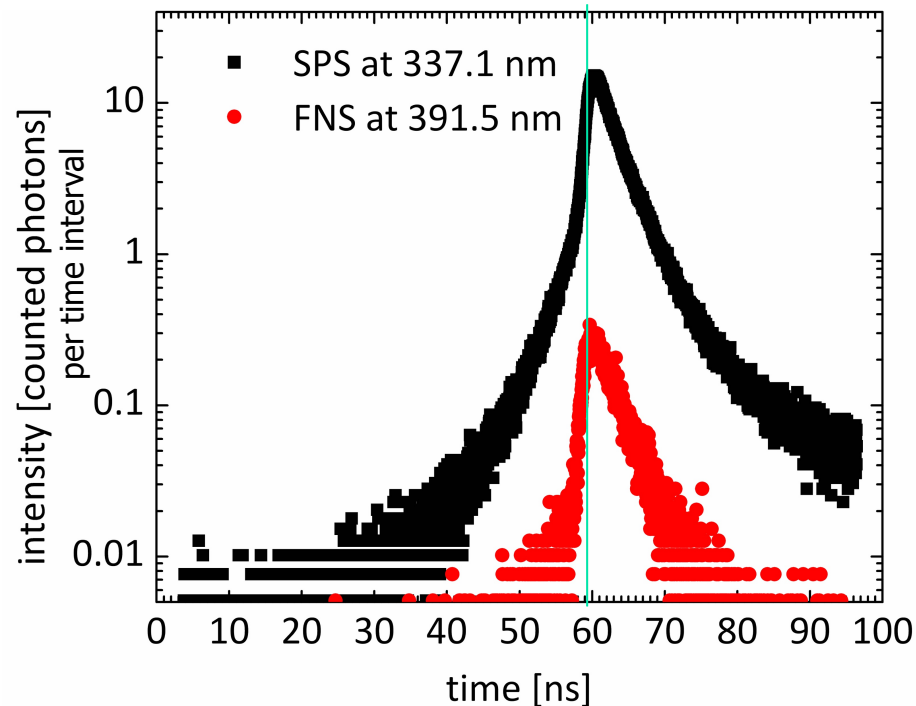
Positive streamer discharges



- **Distinct structure** for the charge transferred by positive streamers
- **Continuous transitions** of transferred charge between subsequent pulses for negative streamers

First results on electric field quantification for discharge at the water interface

Spectrally resolved recordings for positive streamer discharges



- Electric field amplitude over 1000Td (240kV/cm)
- Comparable to the electric fields in other surface barrier discharges
- Necessity to improve the kinetic model – additional quenching processes

Conclusion and Outlook

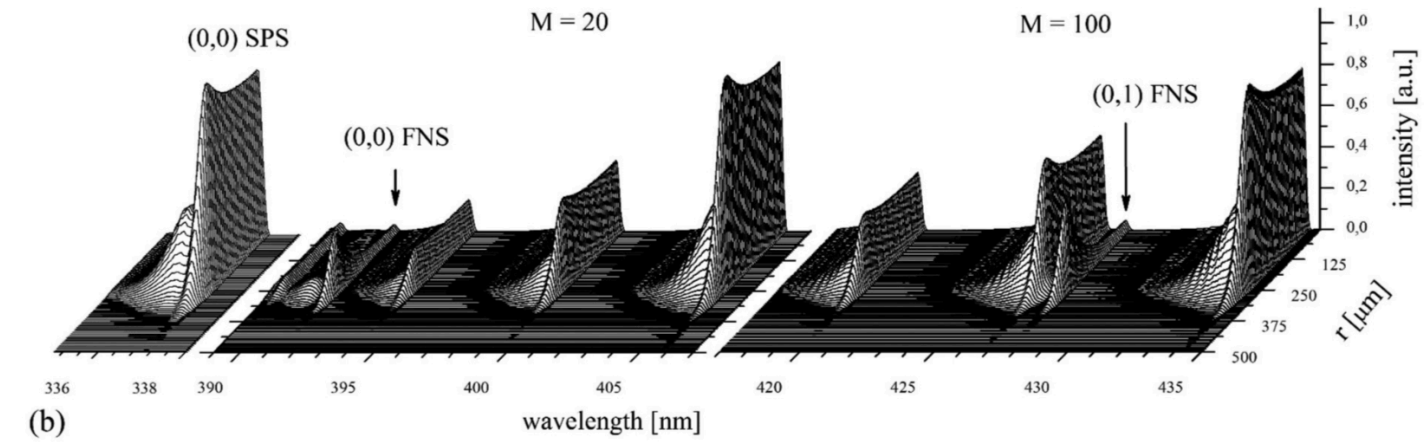
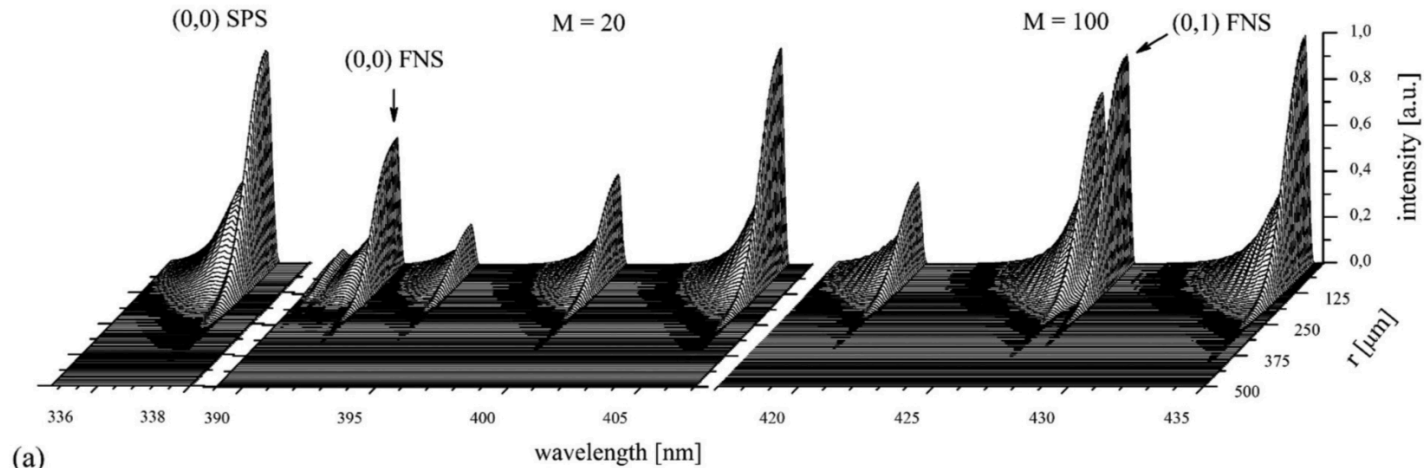
- The **progress in electric field quantification and its uncertainty** was presented for streamer based discharges in atmospheric air
- New technique for **fast and sensitive electrical current measurements** was shown for barrier discharges
- Above mentioned **approaches were applied onto the barrier discharges in contact with water** and preliminary results were shown ... to be finished.

Thank you for your attention!

And many thanks to my colleagues
Zdeněk Bonaventura, Petr Synek, Milan Šimek and others...

This research has been funded by the **Czech Science Foundation project nr.16-19721Y** and also supported by the project CZ.1.05/2.1.00/03.0086 funded by European Regional Development Fund and project LO1411 (NPU I) funded by Ministry of Education Youth and Sports of the Czech Republic.

Optical emission spectroscopy on streamers in air - detailed know-how of streamer head spectrum

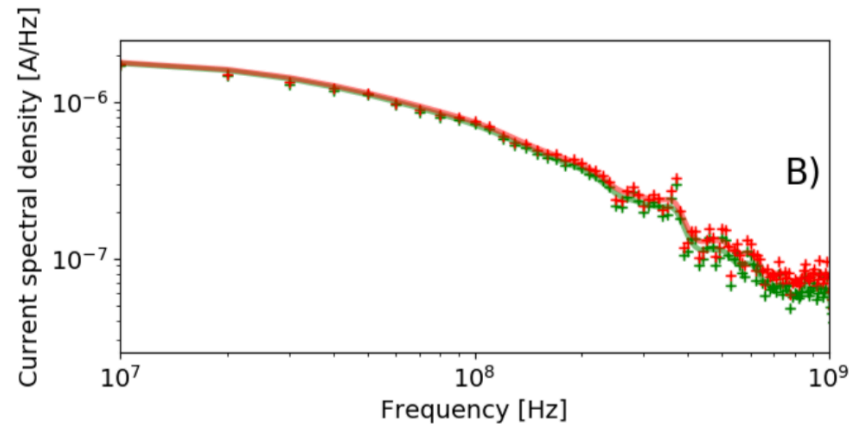
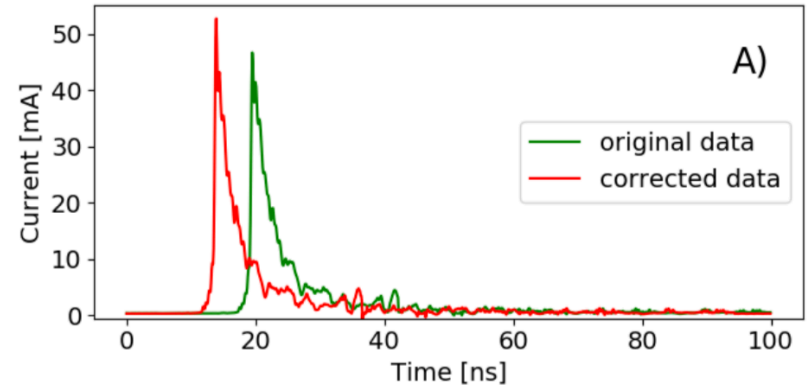


Hoder, Bonaventura, Bourdon et al. 2015 J.Appl.Phys.

Corrected electrical current on the frequency limitations of the current probe and cables

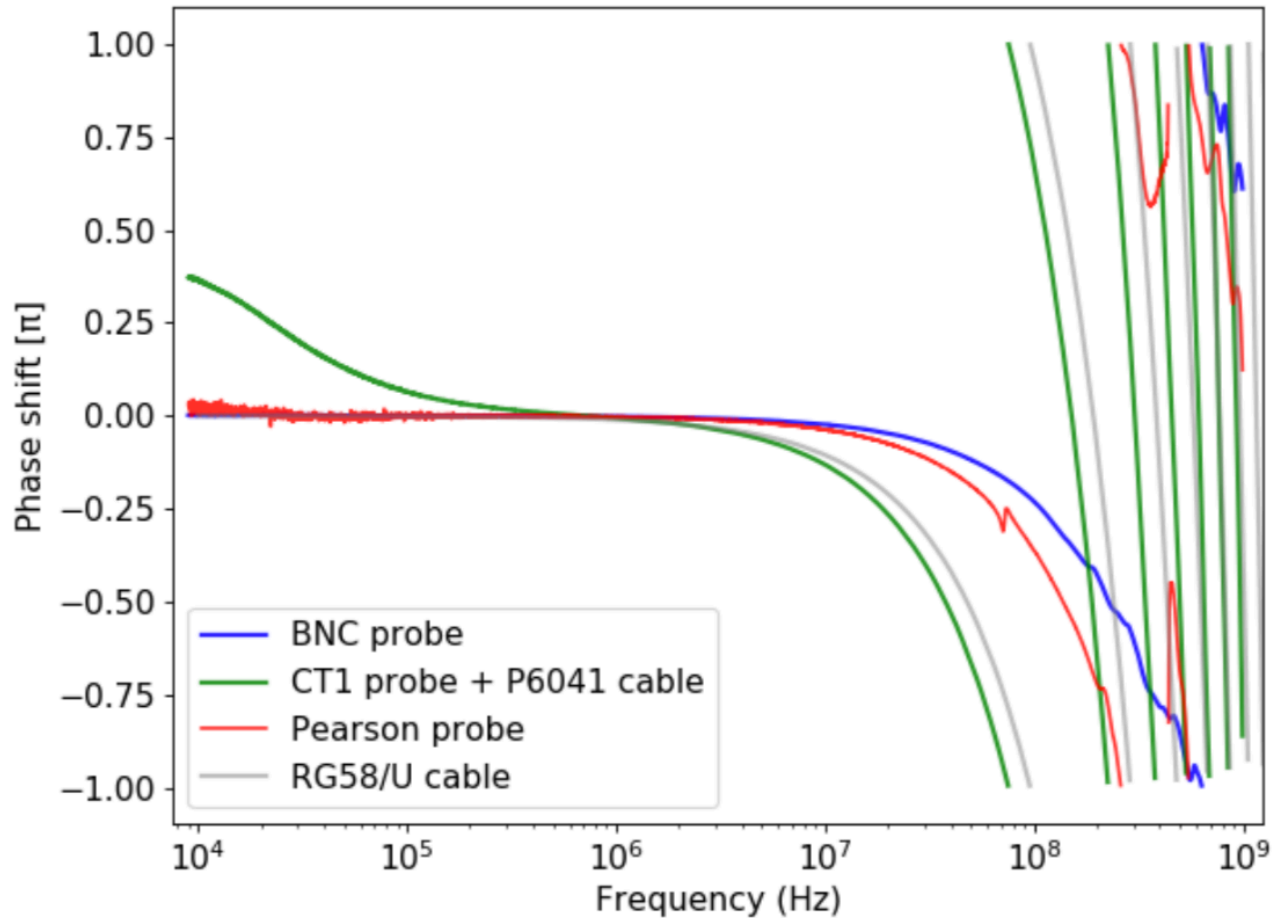
$$FT[f_{real}(\omega)] = \frac{FT[f_{meas}(\omega)]}{\sigma_{cable}(\omega)\sigma_{probe}(\omega)}$$

- Further **increase of the** bandwidth of the probe \gg pulse FWHM 2.0ns
- **Reconstruction of the original current signal** entering the measuring system



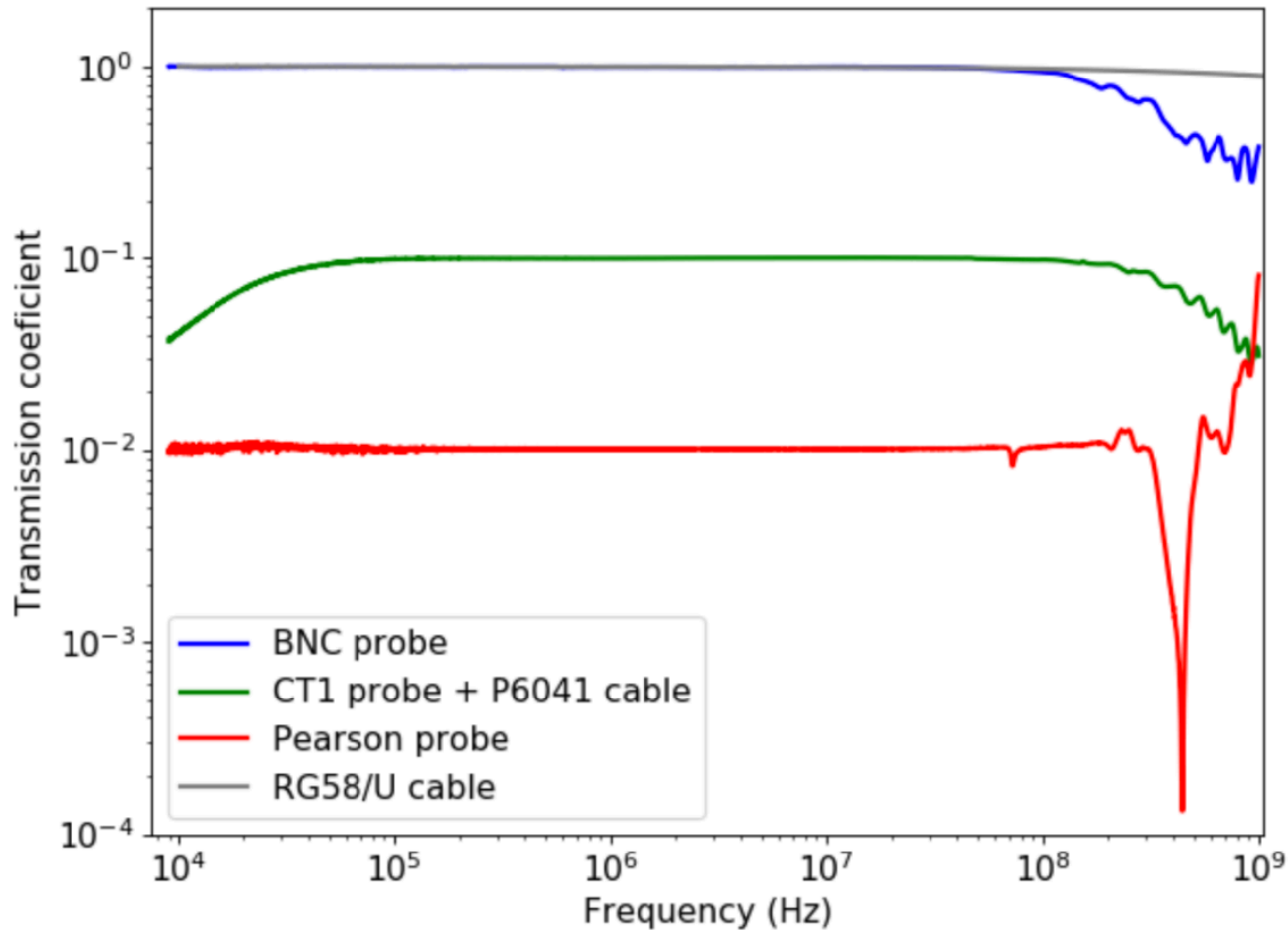
Synek et al. 2018 submitted

Corrected electrical current on the frequency limitations of the current probe and cables



Synek et al. 2018 submitted

Corrected electrical current on the frequency limitations of the current probe and cables

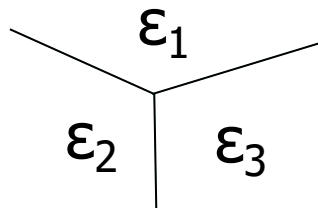


Synek et al. 2018 submitted

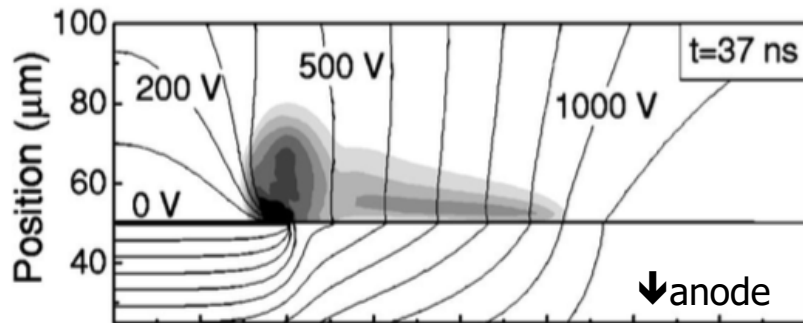
Introduction – triple-junction surface discharges

The special position of triple-junction in discharge physics:

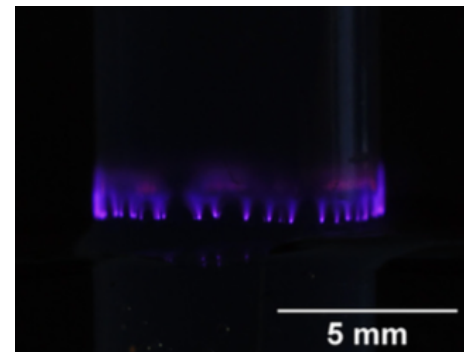
- Typically interface of dielectrics, gas and electrode media (metal stripe or liquid)



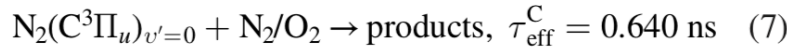
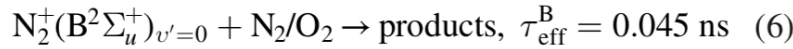
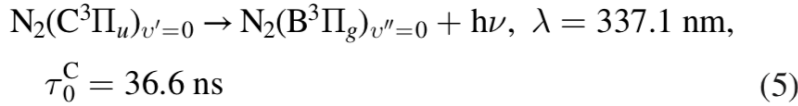
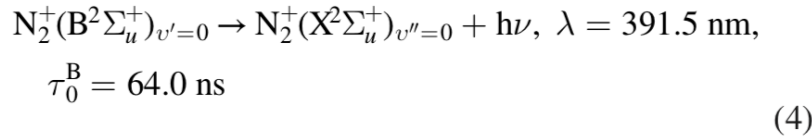
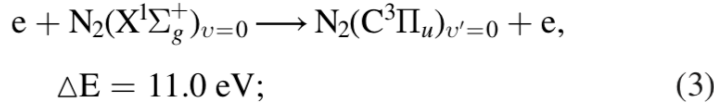
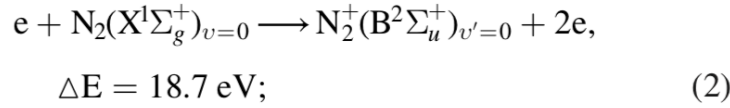
- Surface barrier discharge** for flow control, plasma assisted combustion, polymer treatment and ozone generation – exhibit presence of **strong charge separation in narrow sheath!**



Boeuf and Pitchford 2005 JAP



Pavlinak et al. 2014 APL



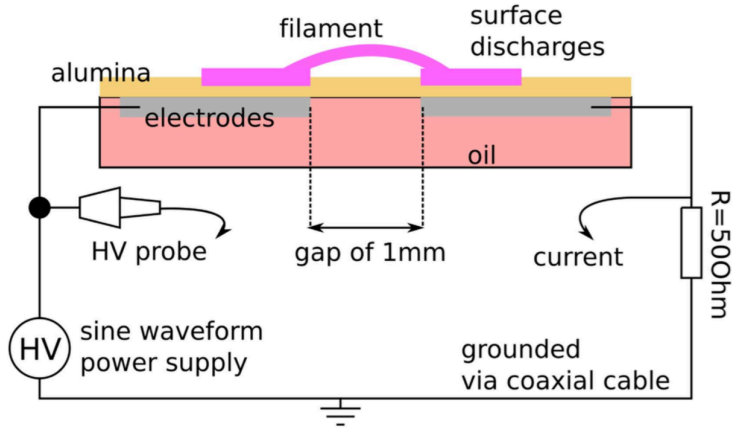
$$\frac{dn_B(x, t)}{dt} = k_B(E/N)n_{N_2}n_e(x, t) - \frac{n_B(x, t)}{\tau_{\text{eff}}^B} \quad (8)$$

$$\frac{dn_C(x, t)}{dt} = k_C(E/N)n_{N_2}n_e(x, t) - \frac{n_C(x, t)}{\tau_{\text{eff}}^C} \quad (9)$$

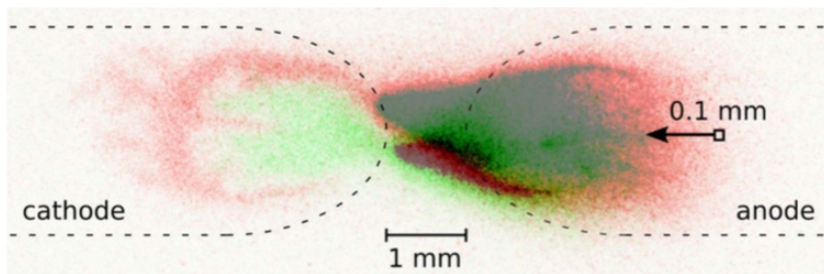
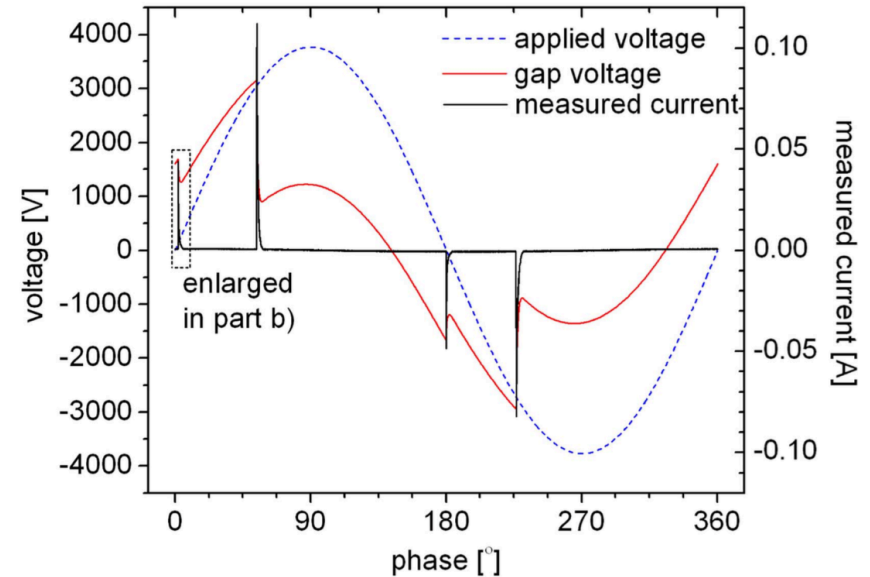
$$\frac{\frac{dI_B(x, t)}{dt} + \frac{I_B(x, t)}{\tau_{\text{eff}}^B}}{\frac{dI_C(x, t)}{dt} + \frac{I_C(x, t)}{\tau_{\text{eff}}^C}} = \frac{k_B(E/N) T_B \tau_{00}^C \lambda_C}{k_C(E/N) T_C \tau_{00}^B \lambda_B} \quad (10)$$

$$\frac{I_B}{I_C} = \frac{k_B(E/N) T_B \tau_{00}^C \lambda_C \tau_{\text{eff}}^B}{k_C(E/N) T_C \tau_{00}^B \lambda_B \tau_{\text{eff}}^C} = R_{\text{FNS/SPS}}(E/N). \quad (11)$$

Recent results in surface barrier discharges in 30kPa air



30 kPa synthetic air
7.2 kVpp sine voltage at 11 kHz



first microdischarge is shown in green color,
the second one in red

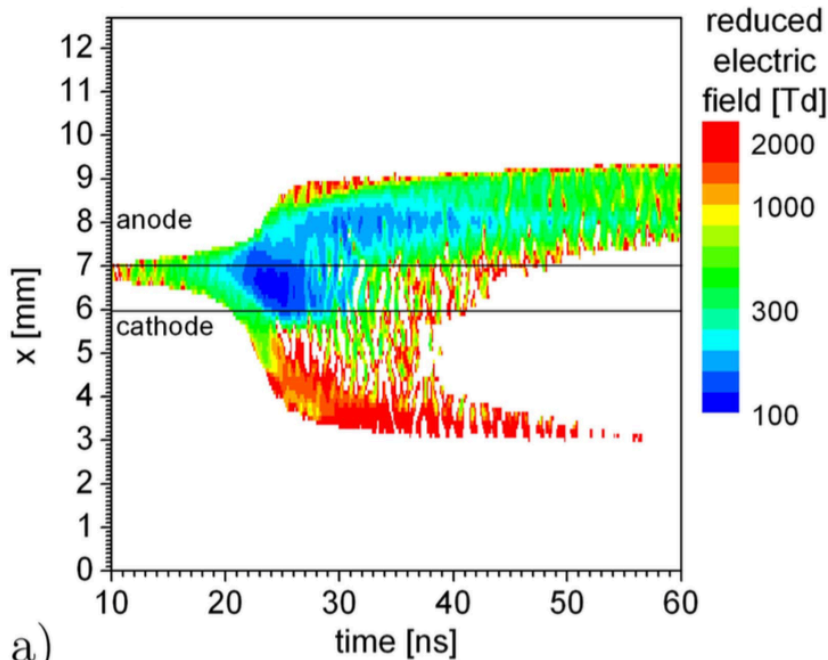
$$U_g(t) = V(t) - \frac{Q(t)}{C_d}$$

access to the mean electric field
in the gap also from electrical measurement

Pipa et al. 2012 Rev. Sci. Instrum.
Hoder et al. 2017 Plasma Phys. Control. Fusion

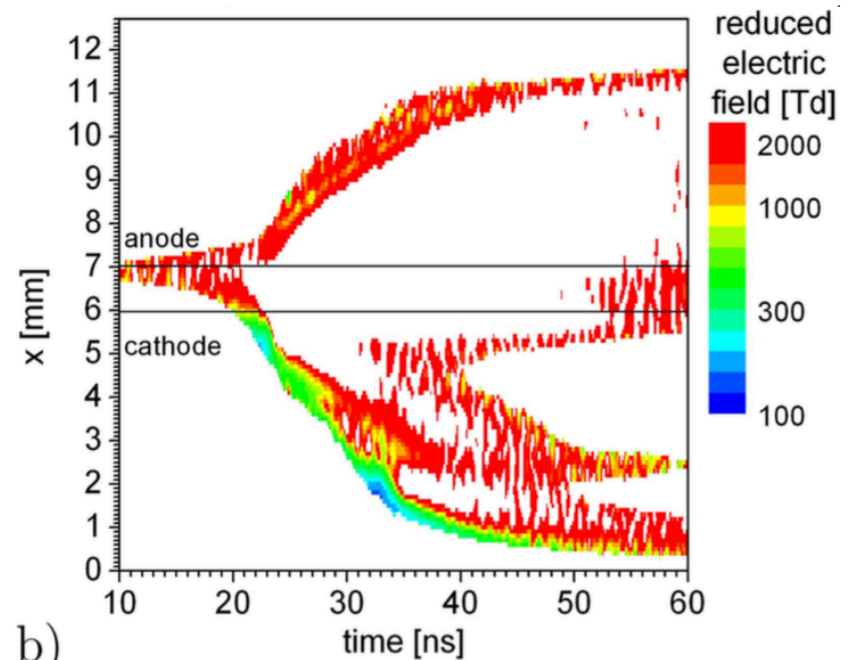
Recent results in surface barrier discharges in air

first microdischarge



a)

second microdischarge



b)

- E/N development within two subsequent microdischarges mutually affected by surface charge
- **Surface streamers of 1200Td** (similar value as in recent hybrid models) and lower for propagation on the contact-line with previously deposited charge

Hoder et al. 2017 Plasma Phys. Control. Fusion
Babaeva et al. 2016 Plasma Sources Sci. Technol.

Nanosecond discharges and laser diagnostics

OPEN ACCESS

IOP Publishing

J. Phys. D: Appl. Phys. **50** (2017) 14LT01 (7pp)

Letter

Ultrafast laser-collision-induced fluorescence in atmospheric pressure plasma

E V Barnat and A Fierro

IOP Publishing

J. Phys. D: Appl. Phys. **50** (2017) 015204 (17pp)

Femtosecond, two-photon-absorption, laser-induced-fluorescence (fs-TALIF) imaging of atomic hydrogen and oxygen in non-equilibrium plasmas

Jacob B Schmidt¹, Suresh Roy¹, Waruna D Kulatilaka¹, Ivan Shkurenkov², Igor V Adamovich², Walter R Lempert² and James R Gord³

Letters

Journal of Physics D: Applied Physics

<https://doi.org/10.1088/1361-6463/aa5f1e>

IOP Publishing

Plasma Sources Sci. Technol. **26** (2017) 055004 (18pp)

Comparison of femtosecond- and nanosecond-two-photon-absorption laser-induced fluorescence (TALIF) of atomic oxygen in atmospheric-pressure plasmas

Jacob B Schmidt¹, Brian Sands², James Scofield³, James R Gord³ and Suresh Roy¹

IOP Publishing

Plasma Sources Sci. Technol. **25** (2016) 054002 (8pp)

Determination of the electric field strength of filamentary DBDs by CARS-based four-wave mixing

P Böhm¹, M Kettlitz², R Brandenburg², H Höft² and U Czarnetzki¹

Plasma Sources Science and Technology

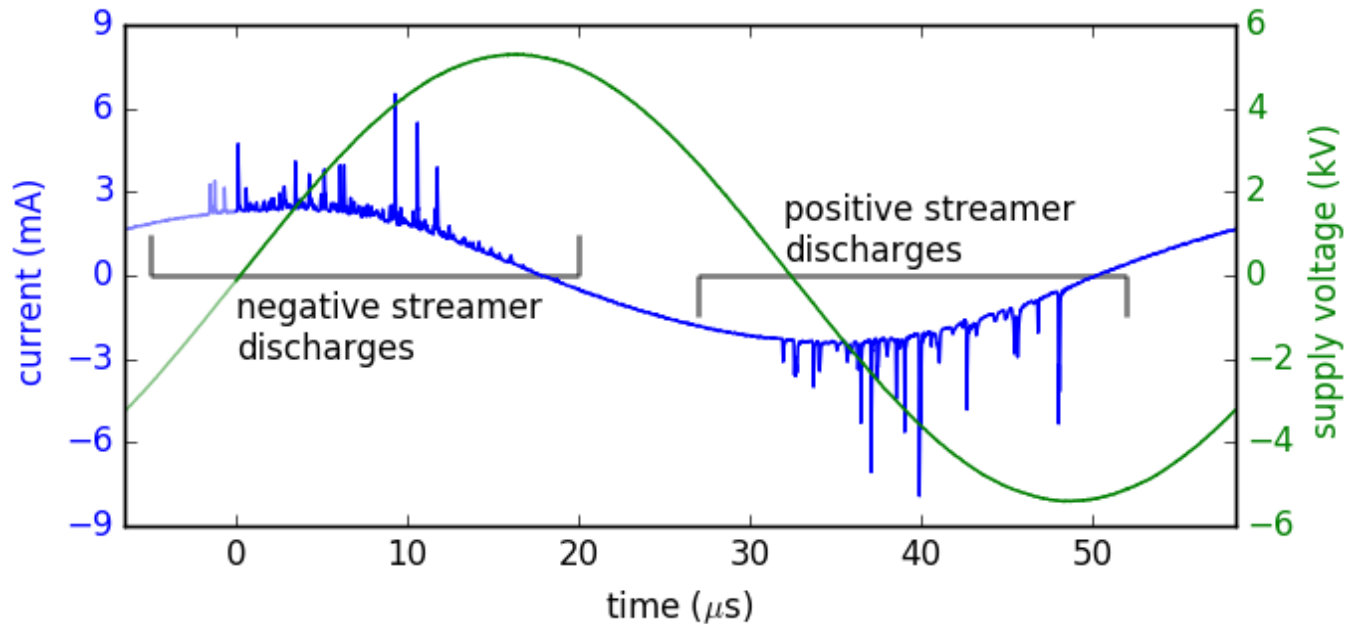
<https://doi.org/10.1088/1361-6595/aa61be>

Plasma Sources Science and Technology

[doi:10.1088/0963-0252/25/5/054002](https://doi.org/10.1088/0963-0252/25/5/054002)

Laser diagnostics offers **direct measurement of the basic plasma parameters** or particle densities. However, the **distortion of the plasma** using higher powers or **unresolved spatial gradients** due to necessary long accumulation paths is still a challenge to solve.

Electric current and voltage characteristic



- Power loss in reactor chamber calculated from electrical measurements data is **1.45 W**.
- Rough estimate of **power consumed in discharges is 0.8 W** while the rest goes to capacitive losses.

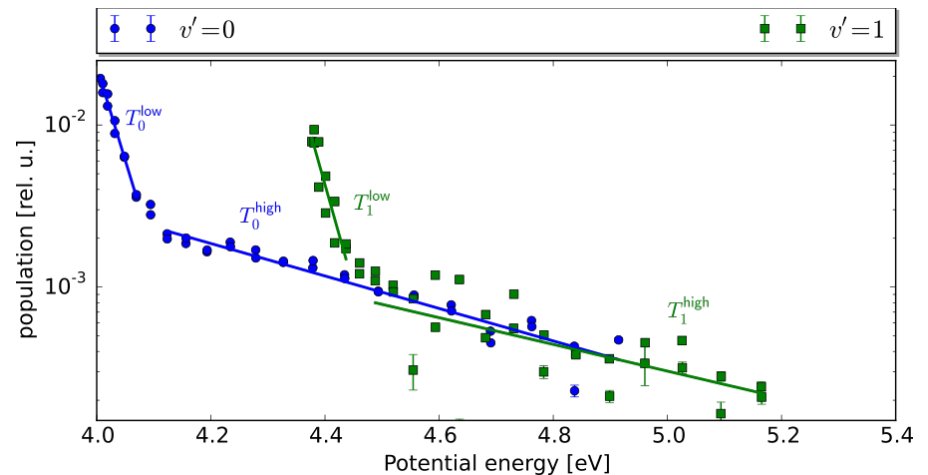
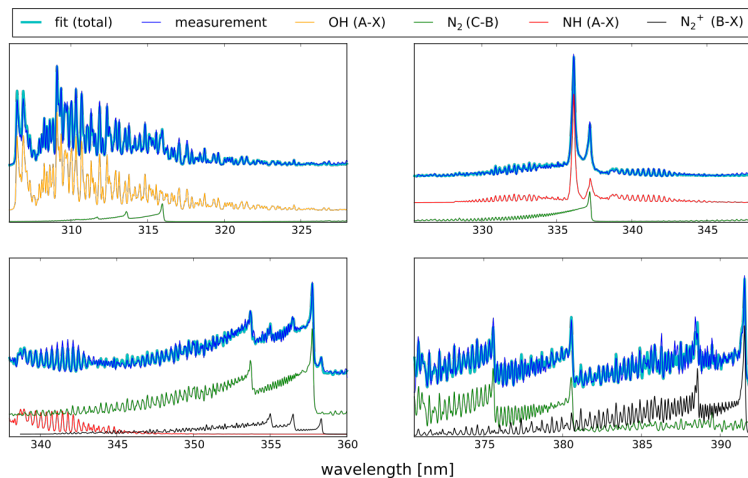
Novel massiveOES free-software

Voráč J, Synek P et al. 2017 PSST

- processed over 5000 spectra of OH, N₂, N₂⁺ and NH molecules

Voráč J, Synek P et al. 2017 JPD:AP

- state-by-state fitting procedure



massiveOES is **FREE** software which allows **batch processing** of large data sets of molecular spectra. Handles **overlapping molecular spectra**. Includes **Boltzmann fit** feature and all essential functions (**linearization**, **line broadening** etc.)

Detailed view of discharges

- **Positive and negative streamer** discharges observed depending on voltage polarity.
- Average **diameter in hundreds of microns**.
- **Cathode spot** was observed for negative streamer discharges at the triple-line
- **Branching phenomena** observed for positive streamers
- In both cases **discharge originates from triple-junction** and propagates along cuvette towards live electrode

