Kinetics of nanosecond discharges at high specific energy release

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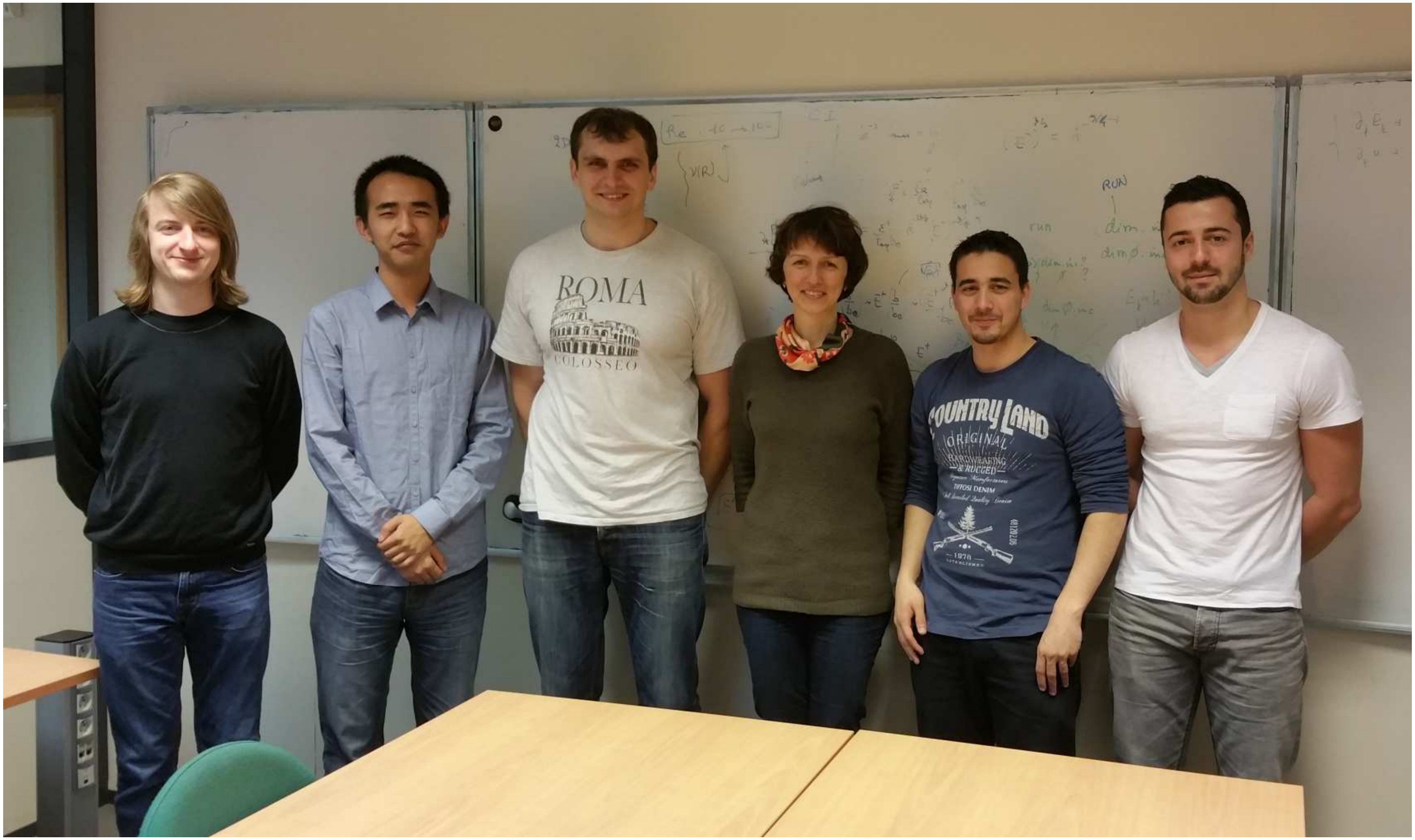
Co-authors and collaborators for this work



Plan of the presentation

I. Introduction

II. Problem formulation

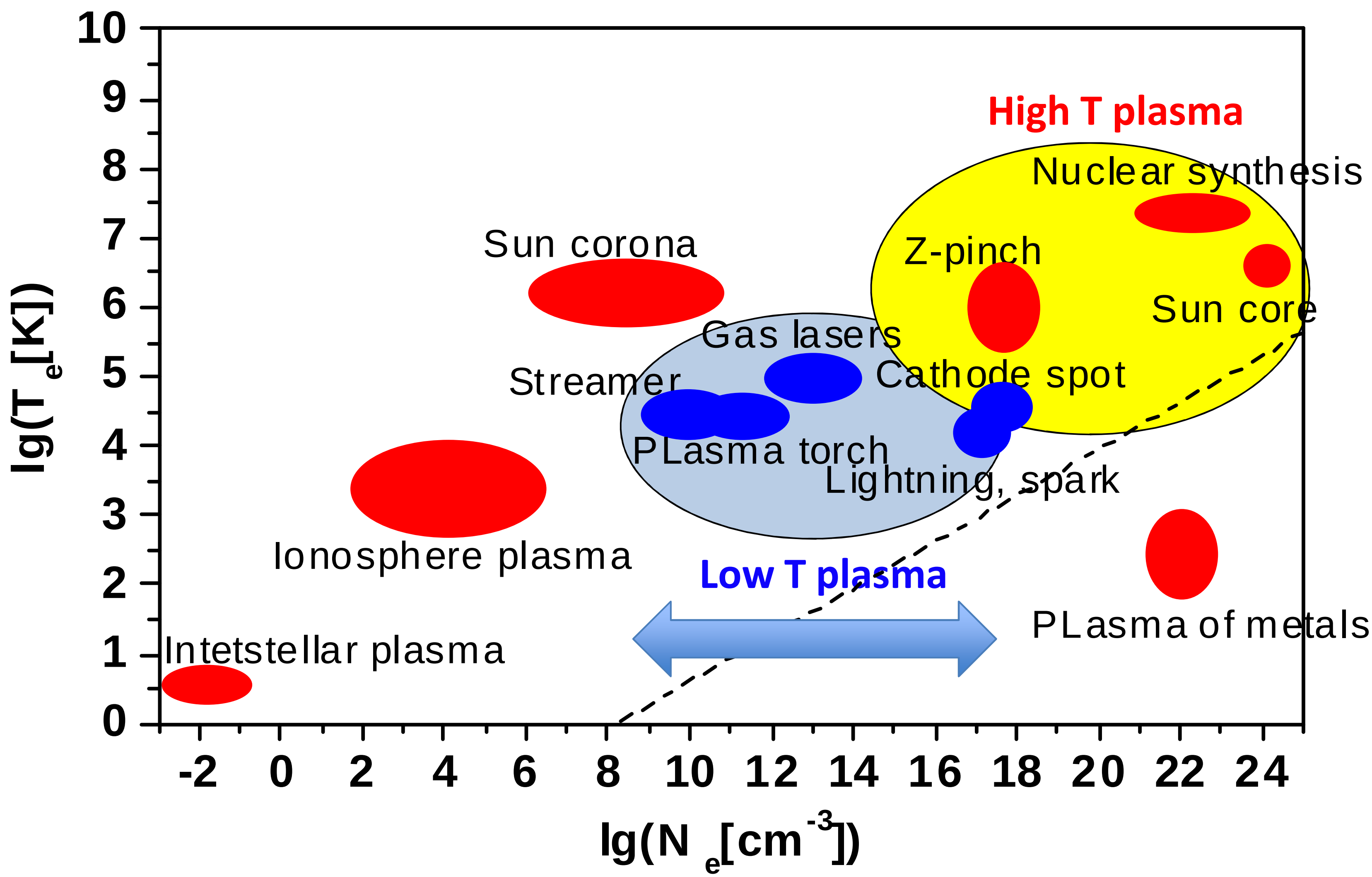
III. Experimental tools

IV. Results and discussion:

-capillary nanosecond discharge at moderate pressure
-surface dielectric barrier discharge (nSDBD) at 1 atm
-surface dielectric barrier discharge at high pressure, filamentation in nanosecond single-shot discharge

V. Conclusion

Low temperature – high temperature plasma: where we are?

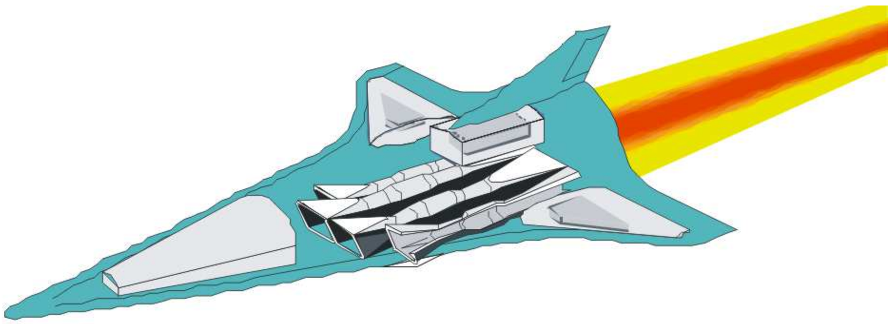


Motivation: plasma-assisted combustion

- Fast flows:

150 ms x 2.5 m/ms (M=8)

L= **375 m**



- To estimate the length:

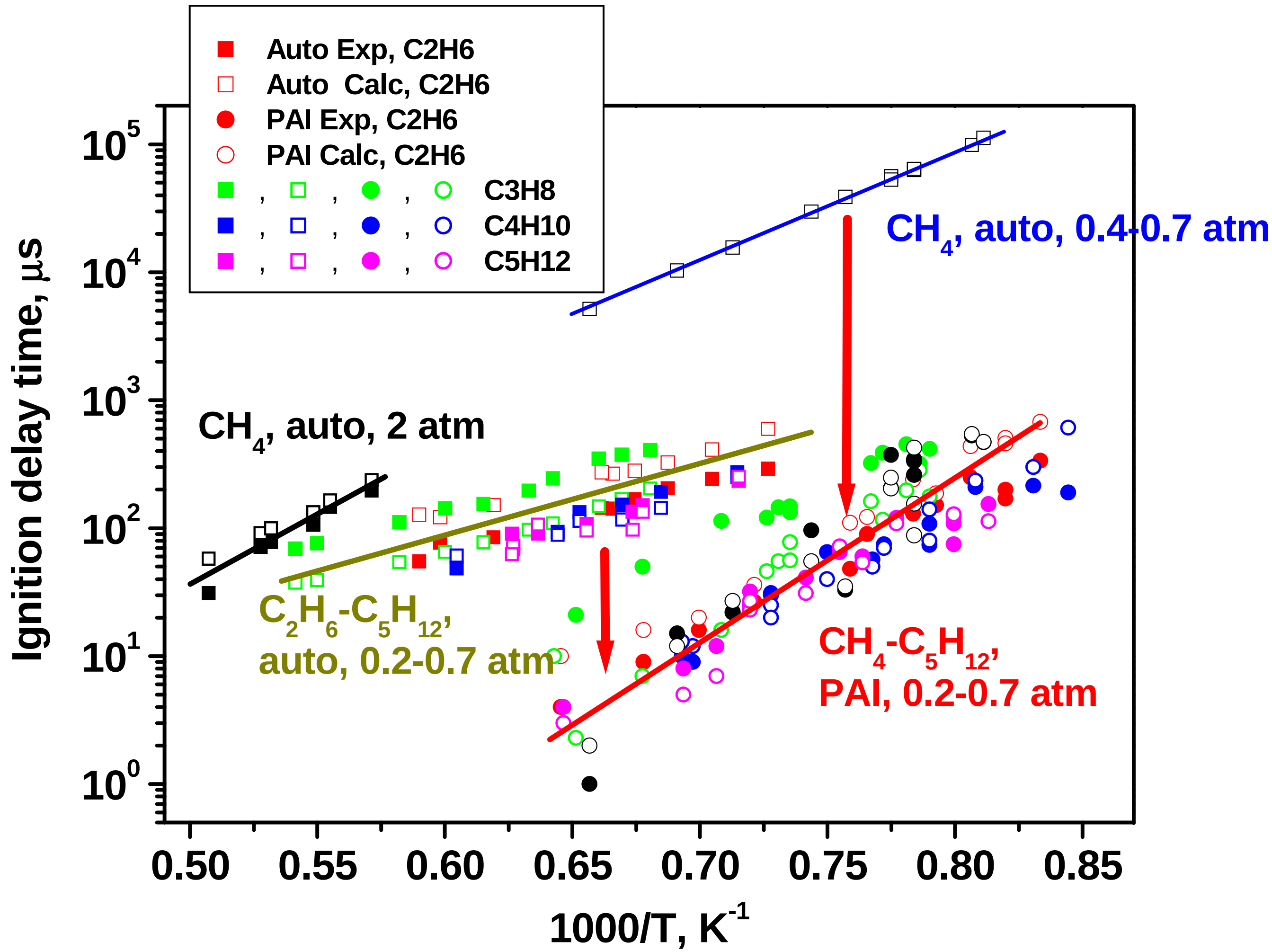
TGV Eurostar:

TransManche Super Train

L_{TGV}=395 m (20 cars)

Need to have shorter ignition delay

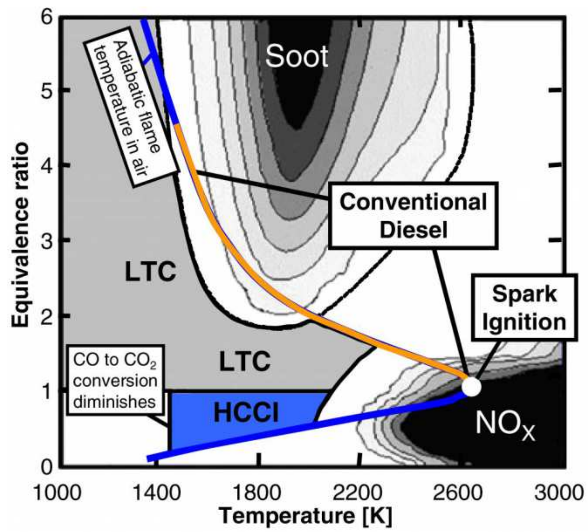
Decrease of ignition delay time: moderate gas densities; uniform ns discharge



N L Aleksandrov, S V Kindysheva, I N Kosarev, S M Starikovskaia, A Yu Starikovskii, *Proc. of Combustion Institute,* 32 (2009) 205-212

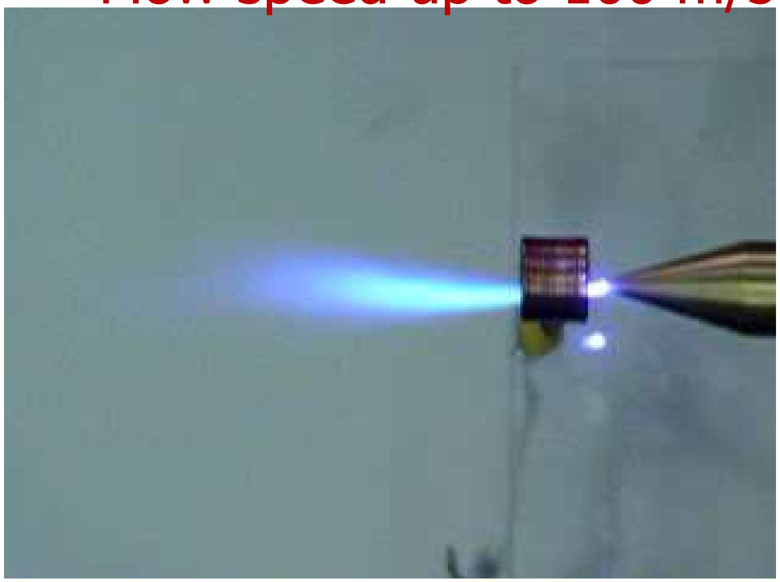
High pressure ignition of combustibe mixtures

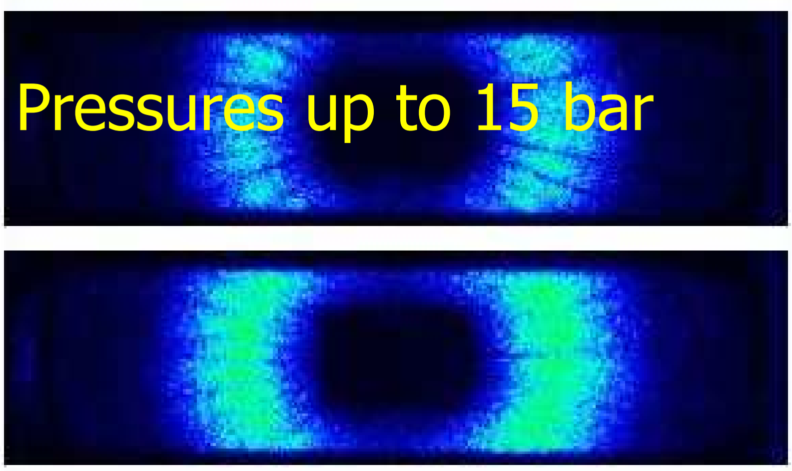
- Ignition of lean mixtures
- Multi-point ignition
- Ignition at lower temperatures
- Ignition of bio-fuels

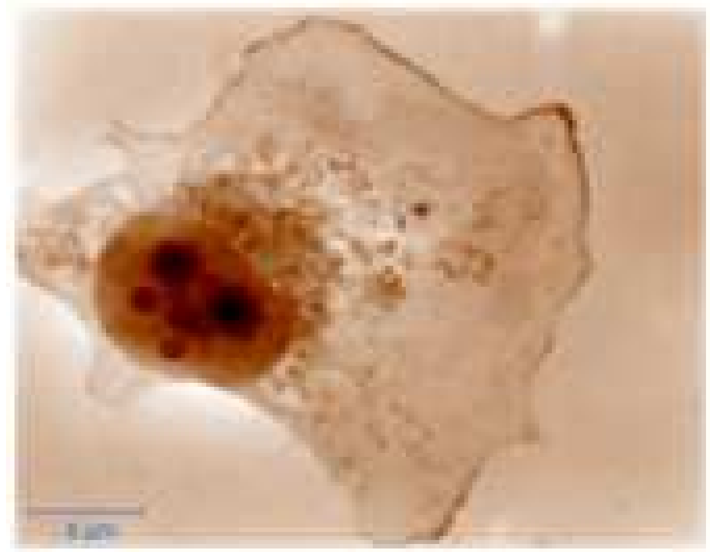


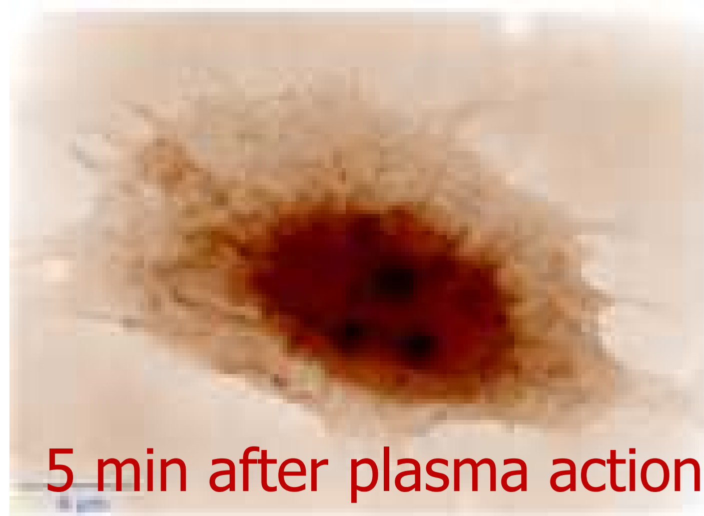
Different potential applications

Flow speed up to 100 m/s

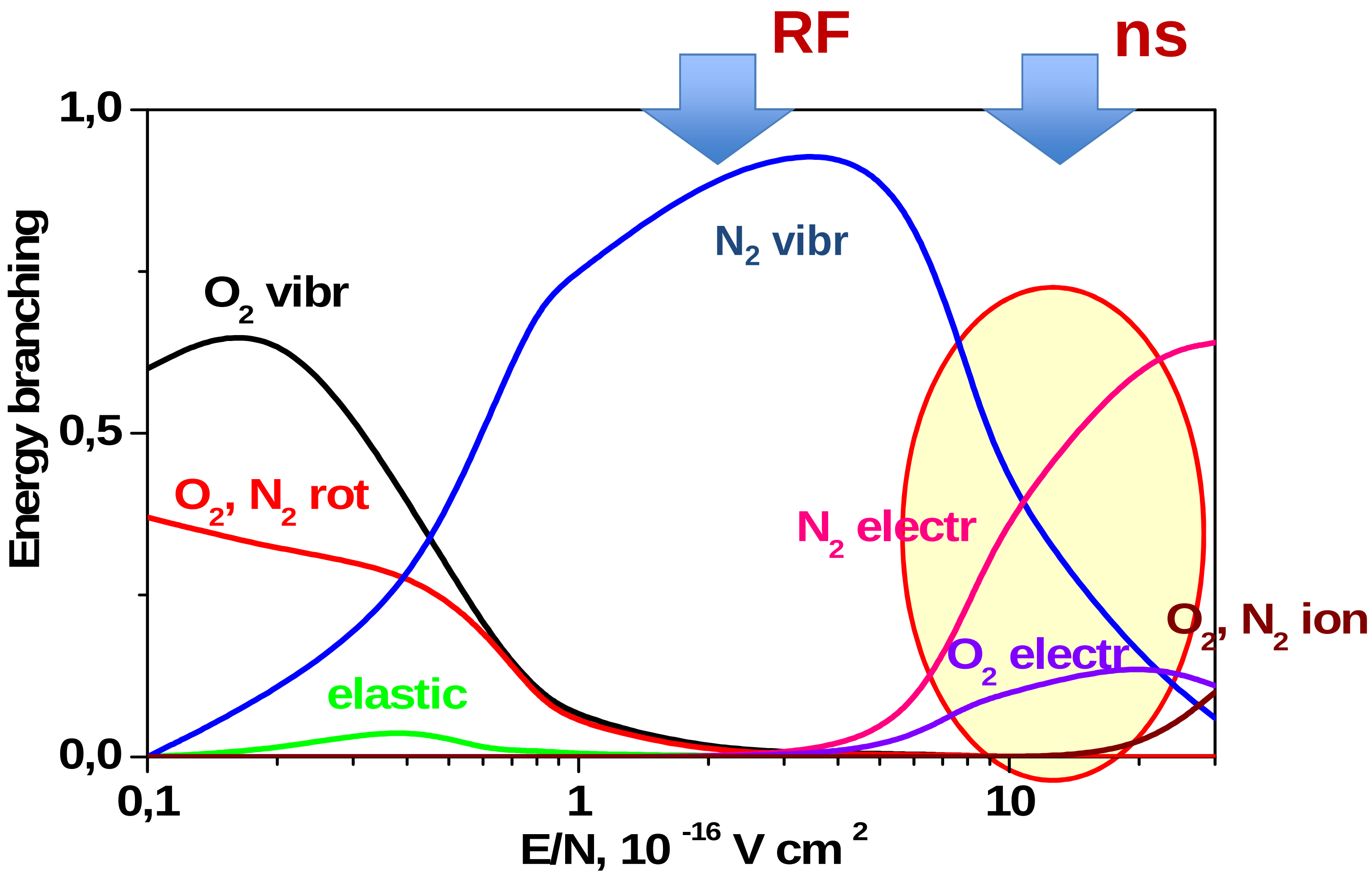








What is peculiar with nanosecond discharges? E-field range



N.L. Aleksandrov, E.E. Son, 1980, in: *Plasma Chemistry,* B.M.Smirnov, ed., Atomizdat Publ., Moscow, V.7, pp. 35-75

Main pathway for initiation [any] chemistry by ns discharges

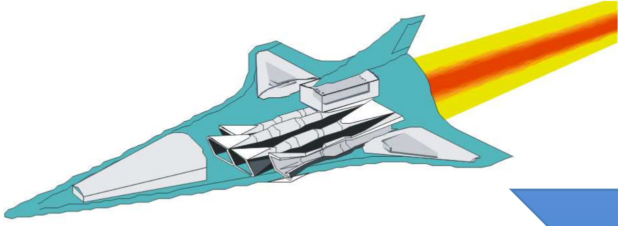
- high **electric fields**

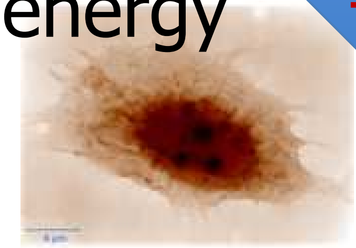
- excitation by electron impact

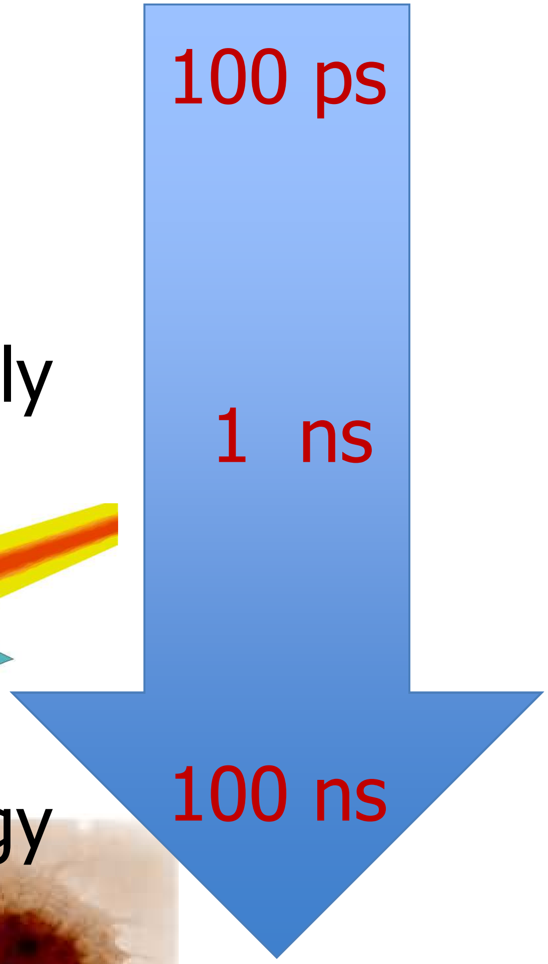
- dominant population of electronically excited levels

- **dissociation**

- relaxation with the rest of the energy going to **heat**







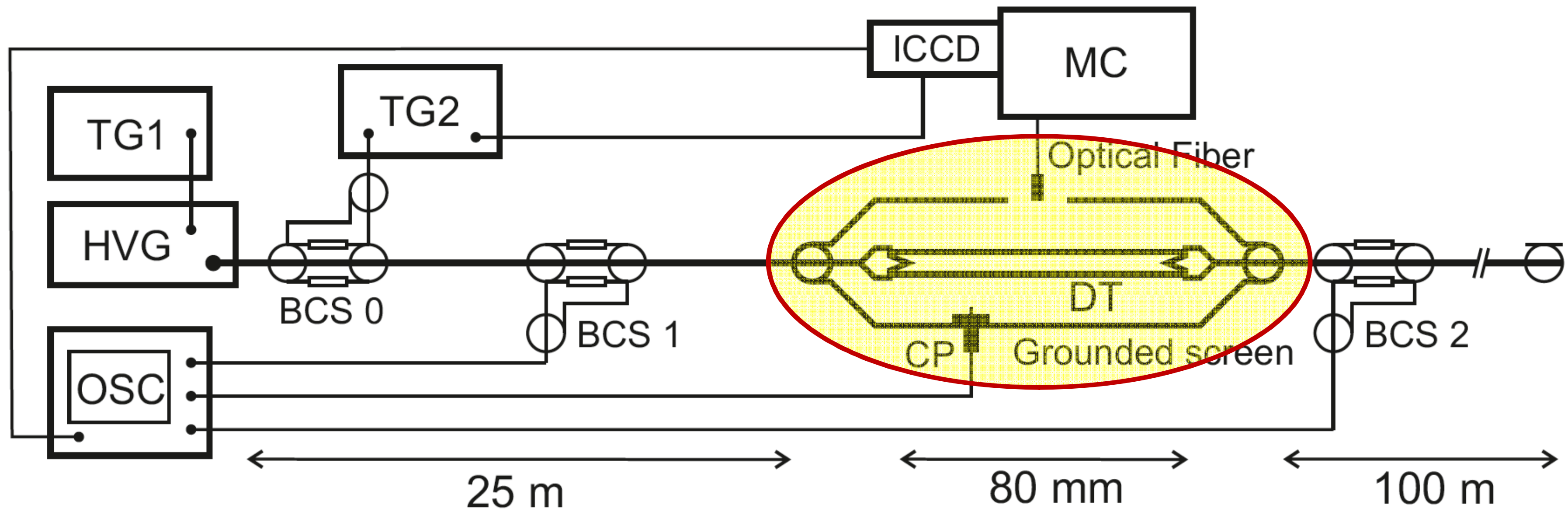
Aim of present work

To underline specific features of pulsed nanosecond discharges under conditions of

- high specific energy density;
- high reduced electric fields.

To specify the most important changes in plasma chemical kinetics at the mentioned conditions

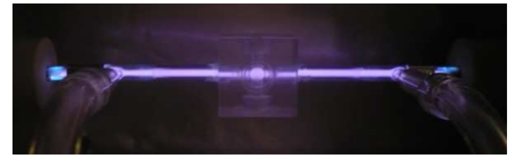
Experimental setup: discharge parameters and spectroscopy



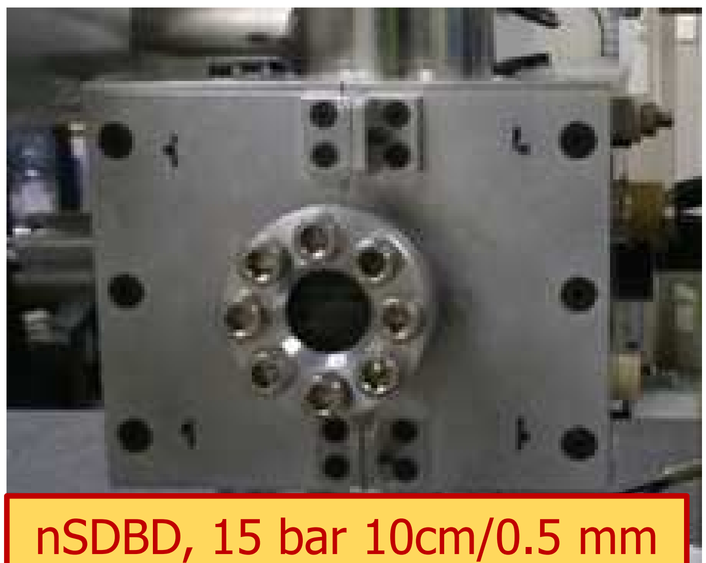
DT - Discharge tube; **BCS** - Back Current Shunt;
TG - Triggering generator; **HVG** - High Voltage Generator;
CP - Capacitive Probe; **ICCD** - Intensified CCD Camera;
OSC - Oscilloscope; **MC** - Monochromator

Typical discharges and discharge cells

FIW, 1-10 Torr,
L=20cm



CD, 20 Torr,
8 cm



nSDBD, 15 bar 10cm/0.5 mm
(T<700 K)

nSDBD, 8 bar
10cm/0.5 mm

Experimental tools

U(t),I(t): calibrated custom made back current shunts;

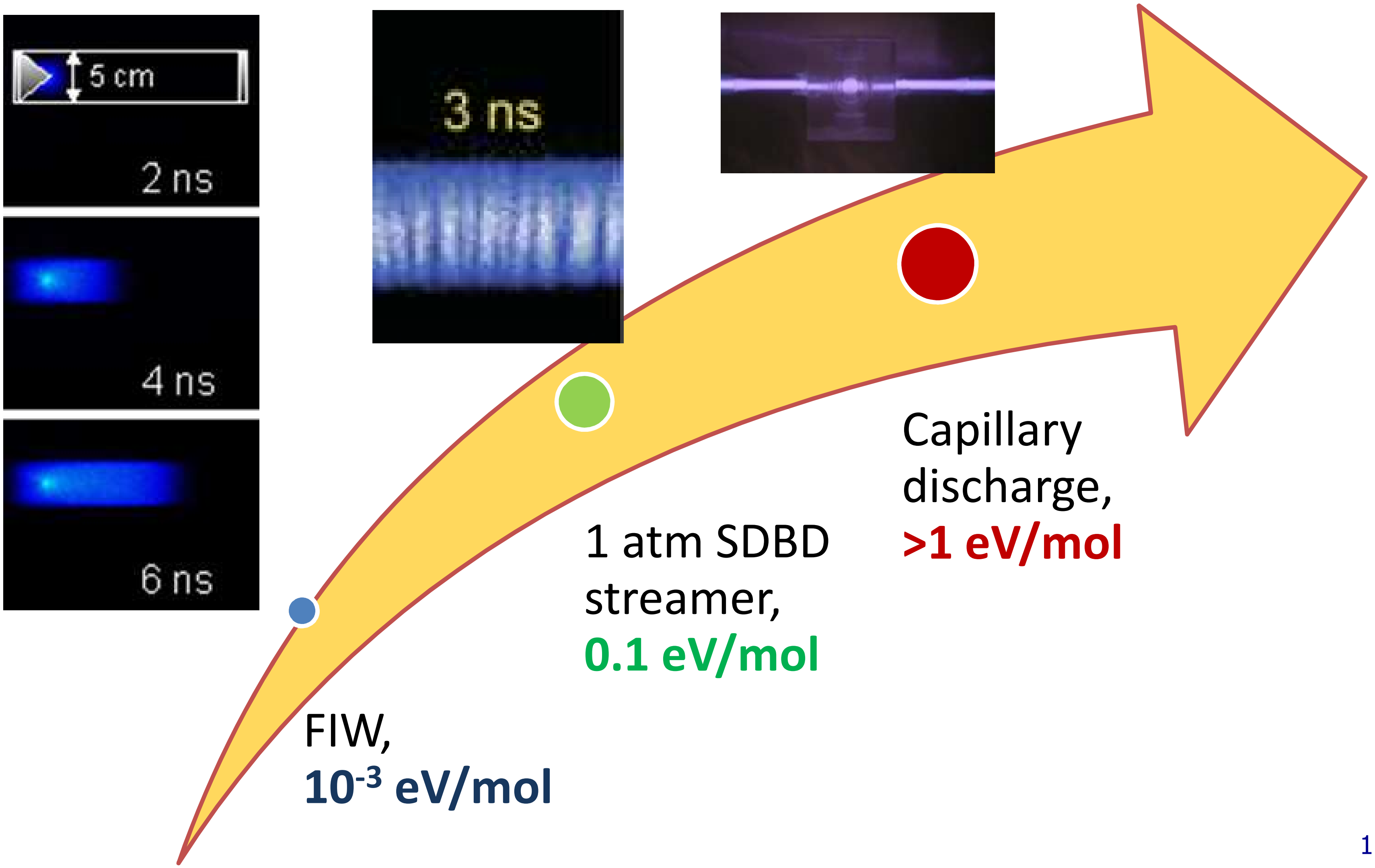
E/N(t,x): calibrated custom made capacitive detectors; emission spectroscopy;

v(t,x), morphology: ICCD (filtered) imaging

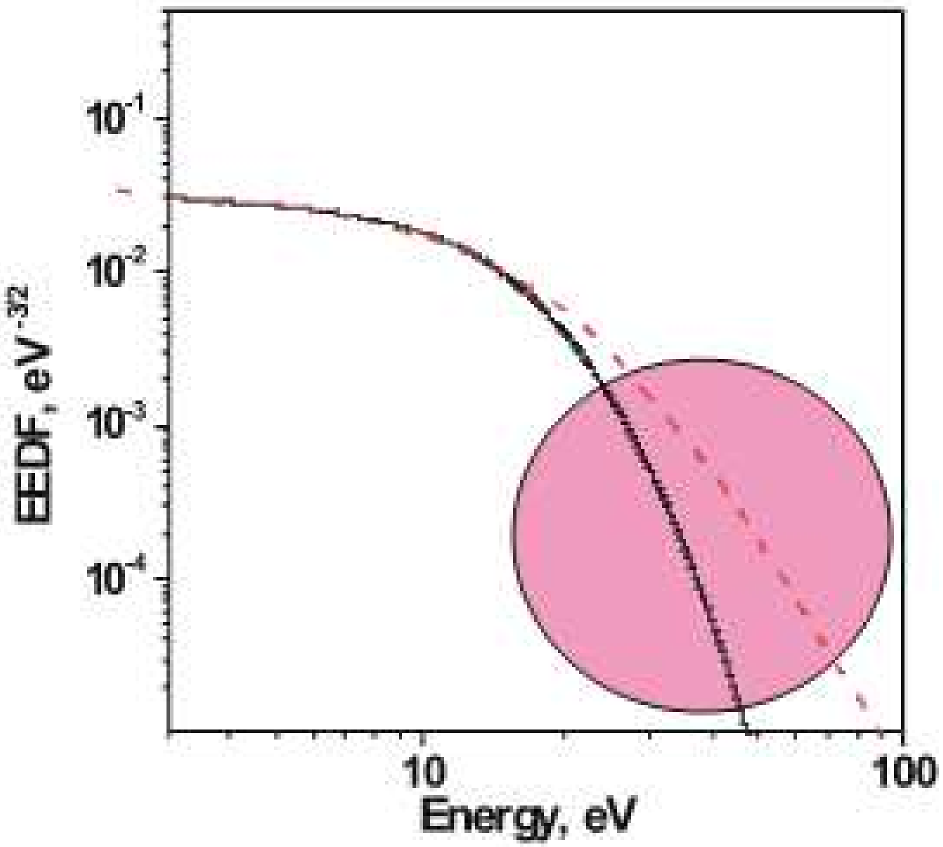
n_e(t,x): electrical current measurements, Stark broadening

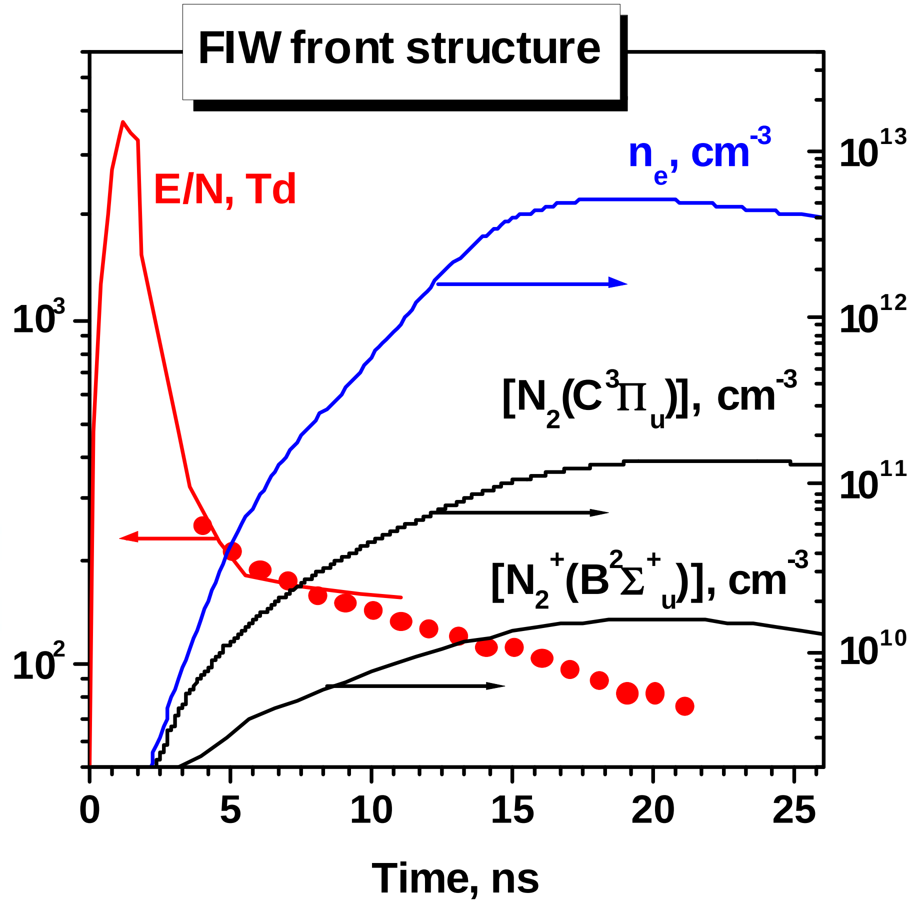
[O](t): actinometry, TALIF

Specific deposited energy, eV/mol



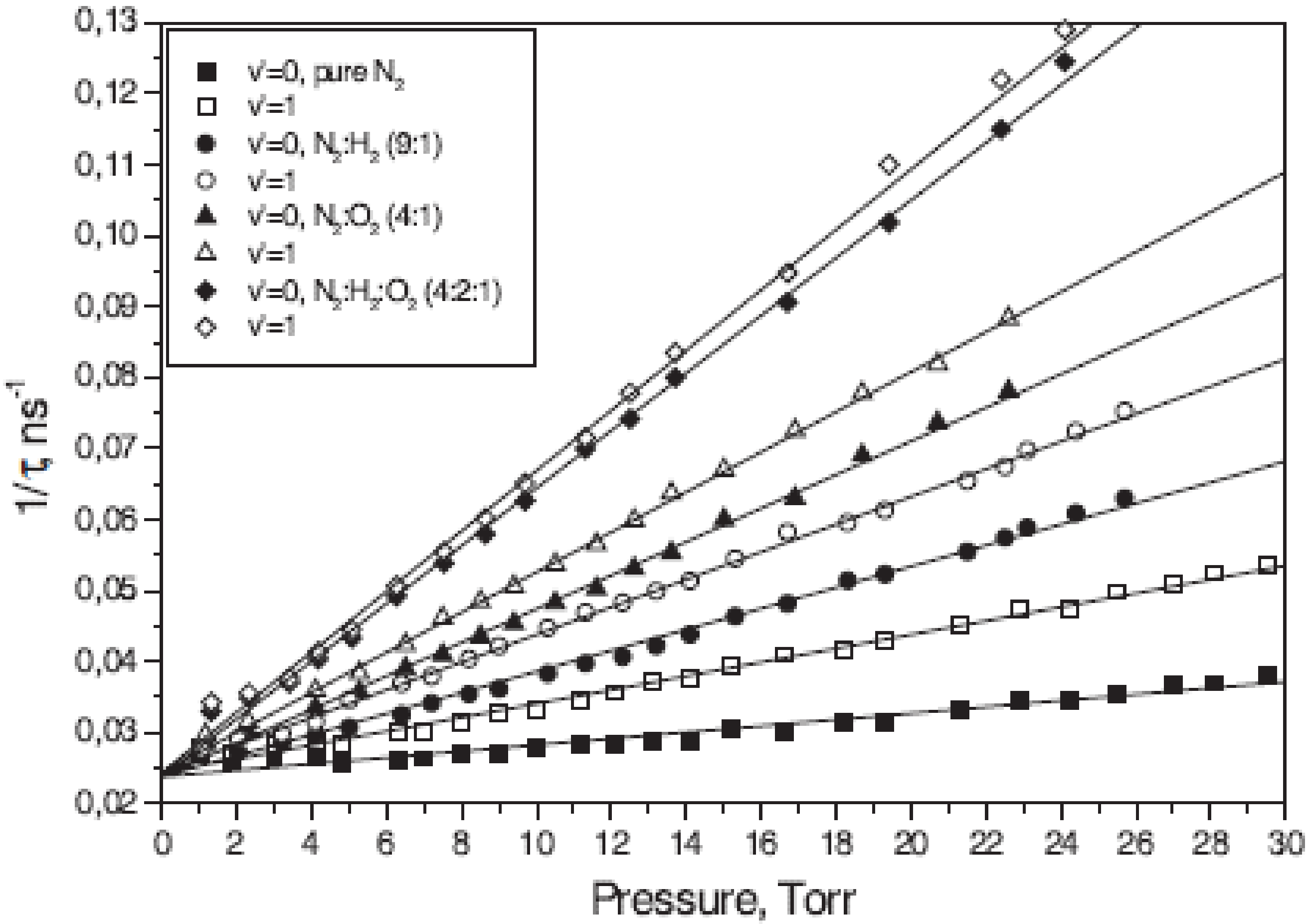
Scientific elegance: "diluted" chemistry in high fields at nanoseconds





S.V.Pancheshnyi et al *J. Phys. D: Appl. Phys,* 1999, 32 2219

Example of application: $N_2(C^3\Pi_u)$ quenching rate measurements



S.V.Pancheshnyi et al *Chem. Phys.*, 2000, 262, 349-357

Fast ionization waves (U=+10 kV in the cable, FWHM=30 ns)

Discharge in air at 1-10 Torr ($N=10^{17}$ cm^{-3}):

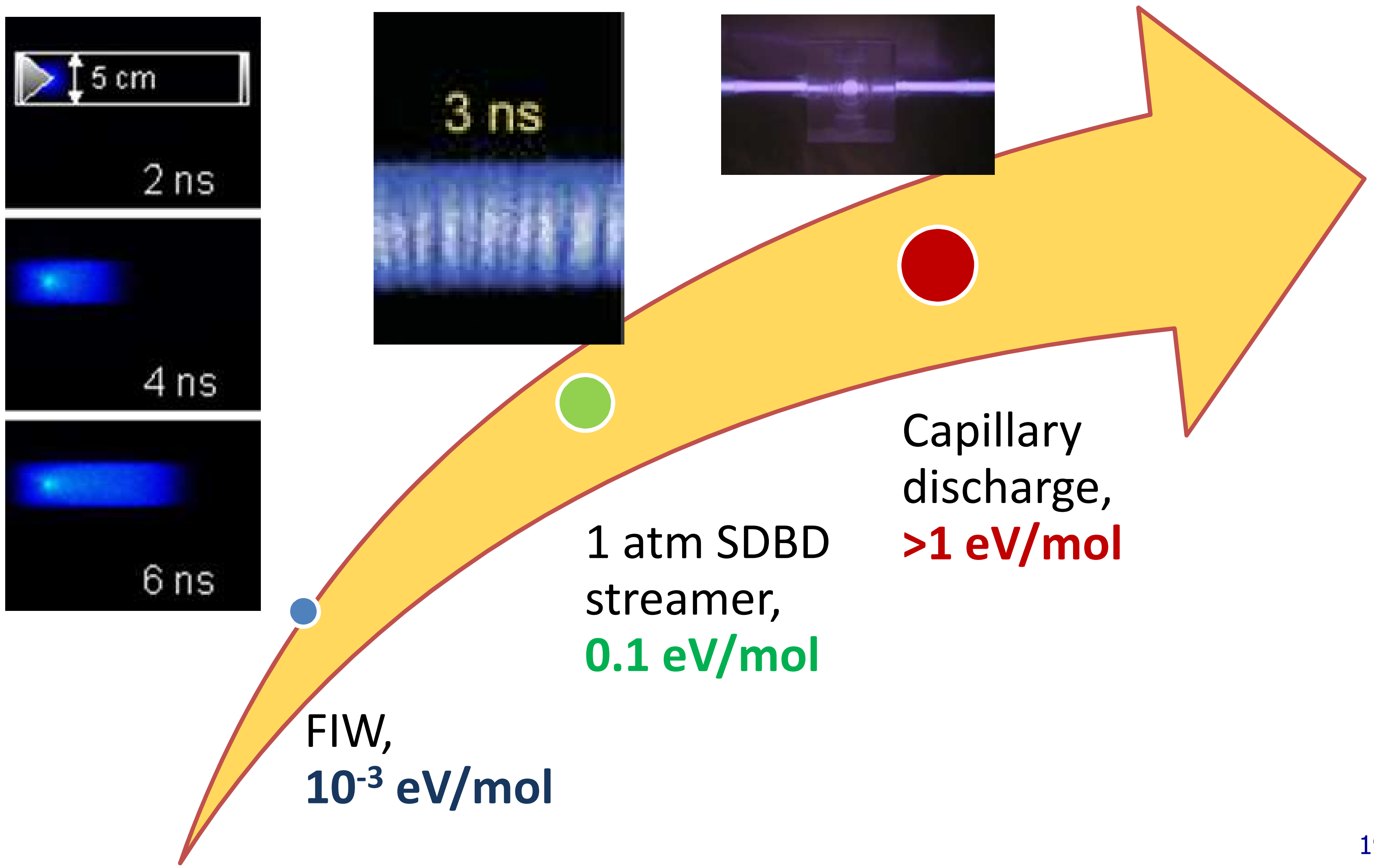
I(t): 60 A (3 A/cm^2);

v(t,x): 50 mm/ns;

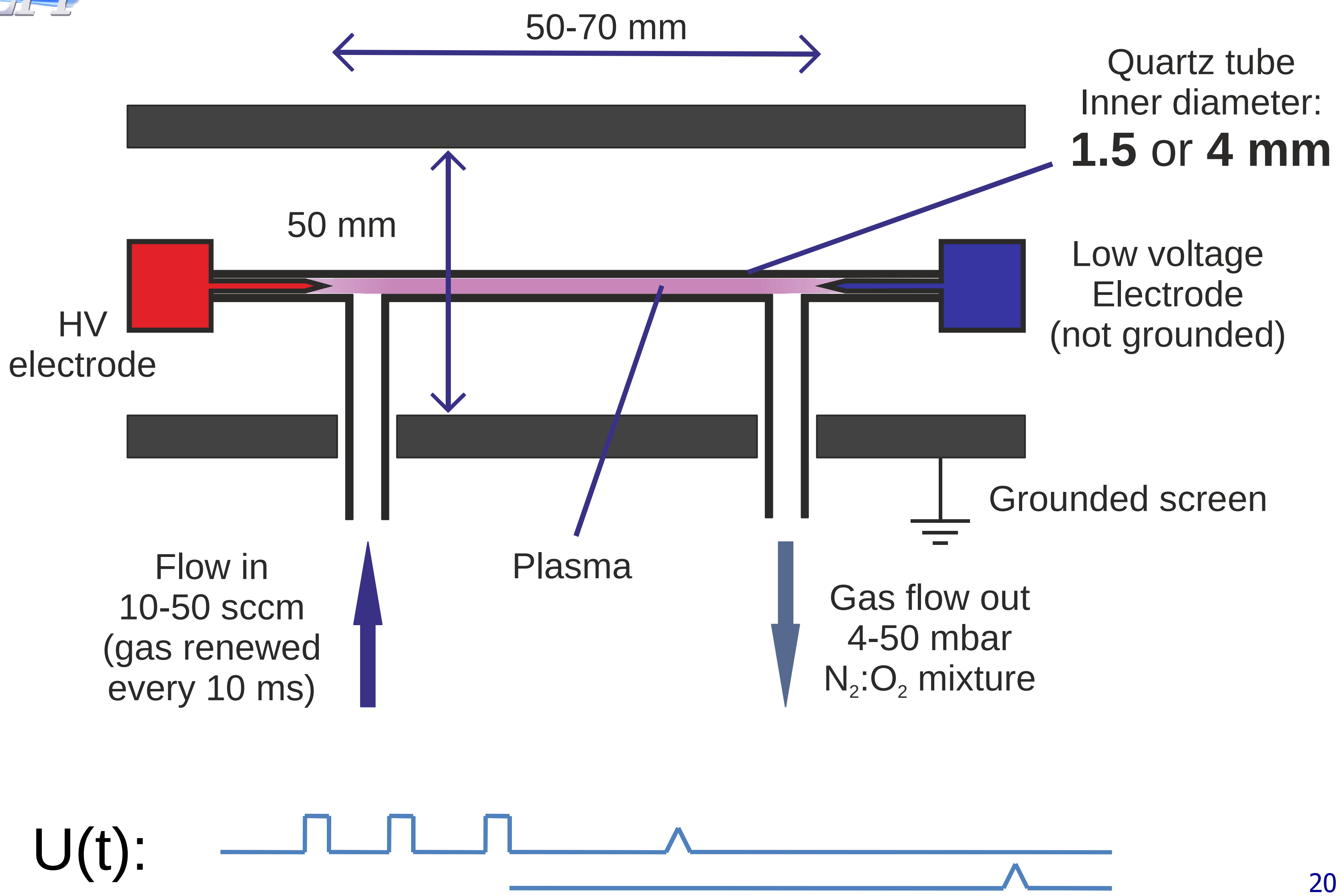
E/N(t,x): 1-5 kTd in the front and 100-300 Td in the region of energy deposition after the front;

$n_e(t,x)$: 10^{12} cm^{-3}

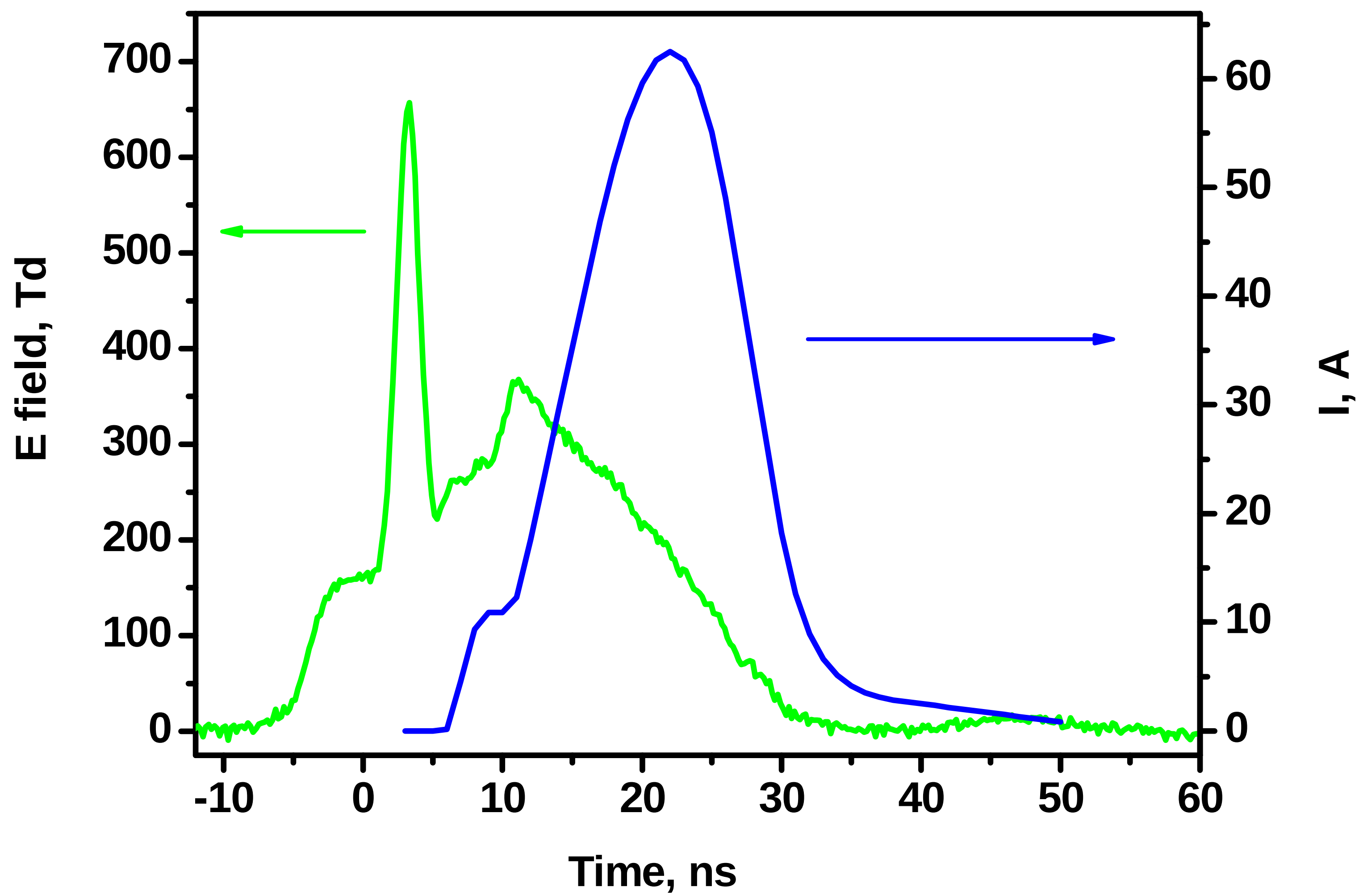
Scale of specific deposited energy



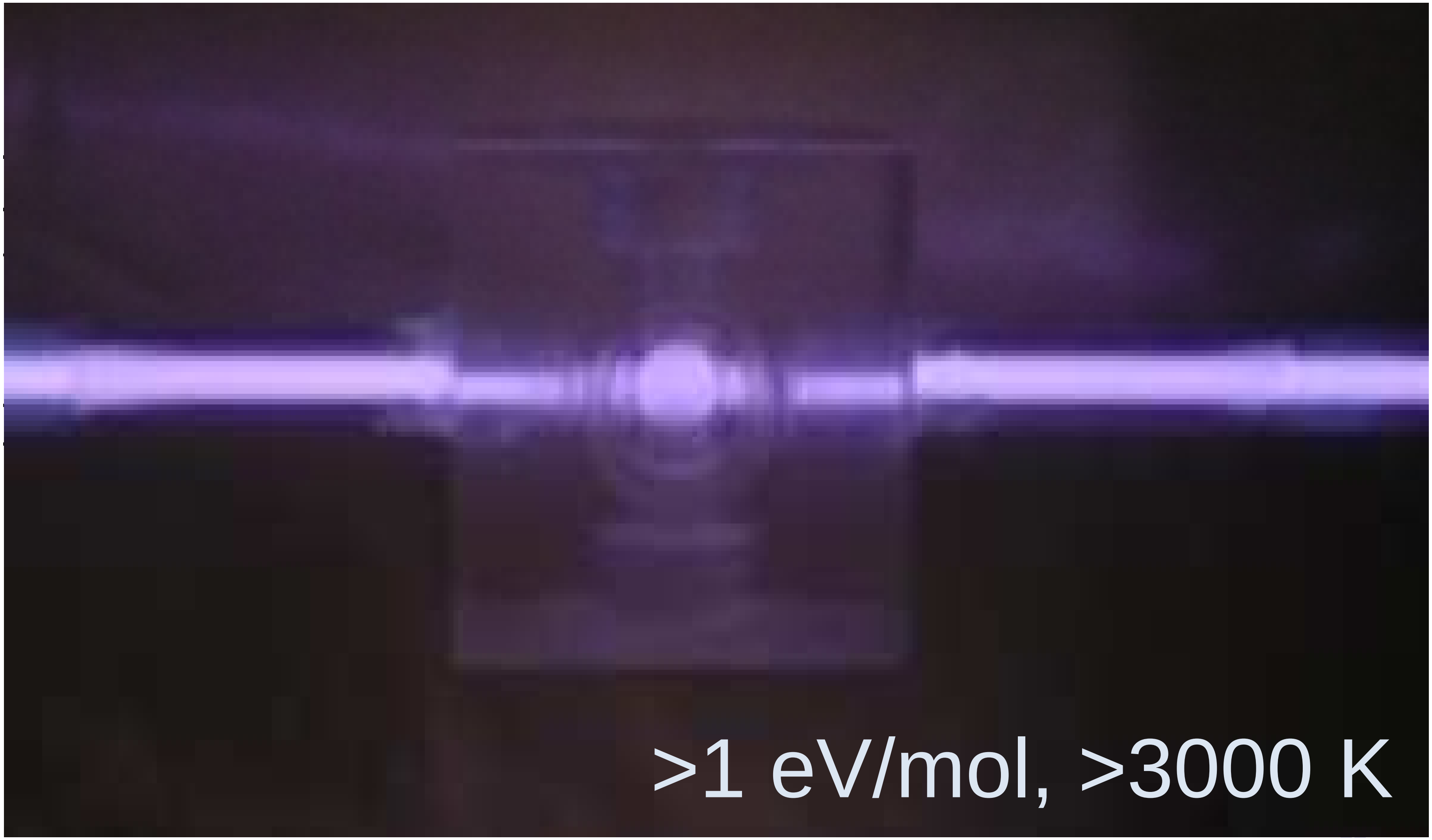
Capillary nanosecond discharge



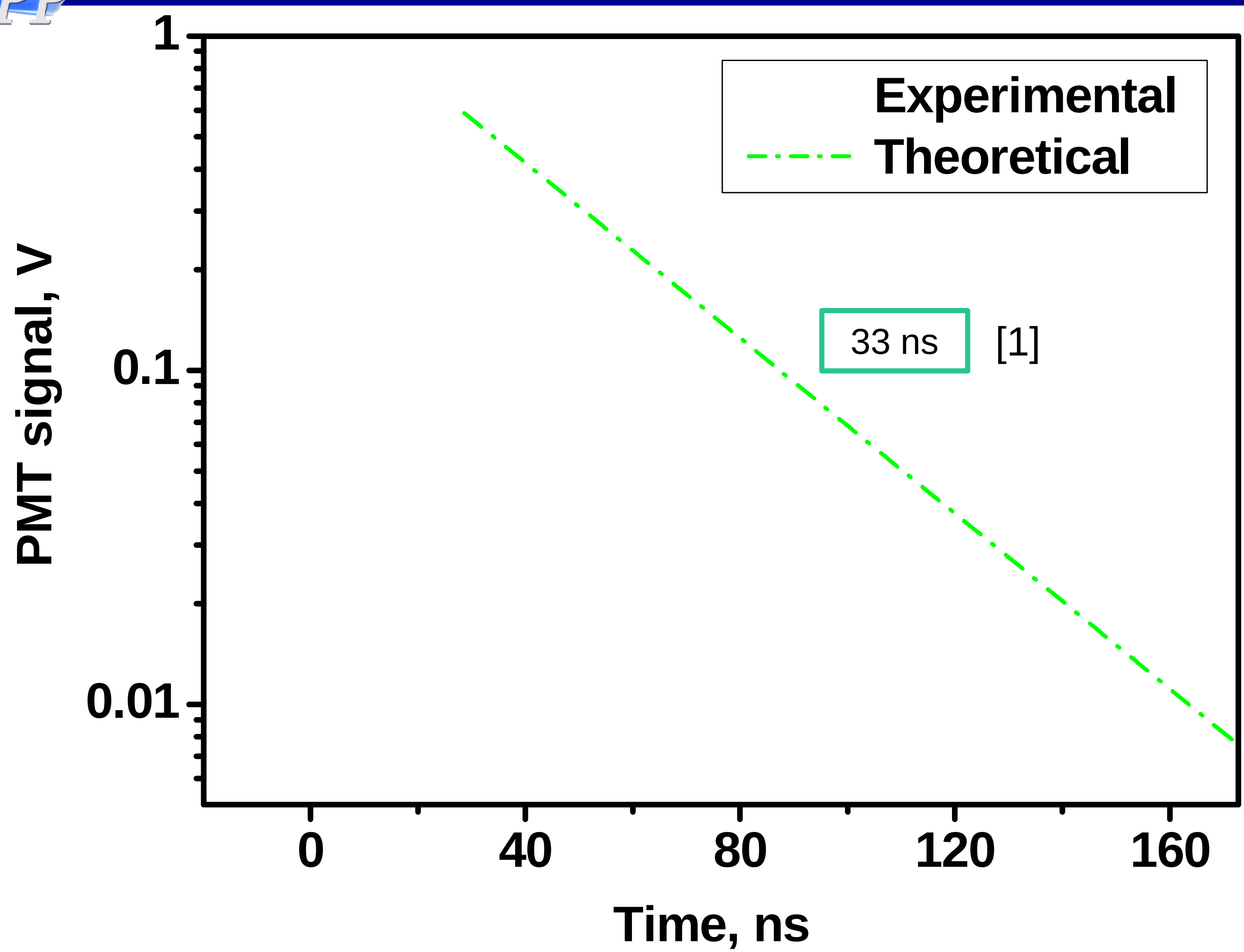
Electric field and current waveforms



Specific energy and gas heating

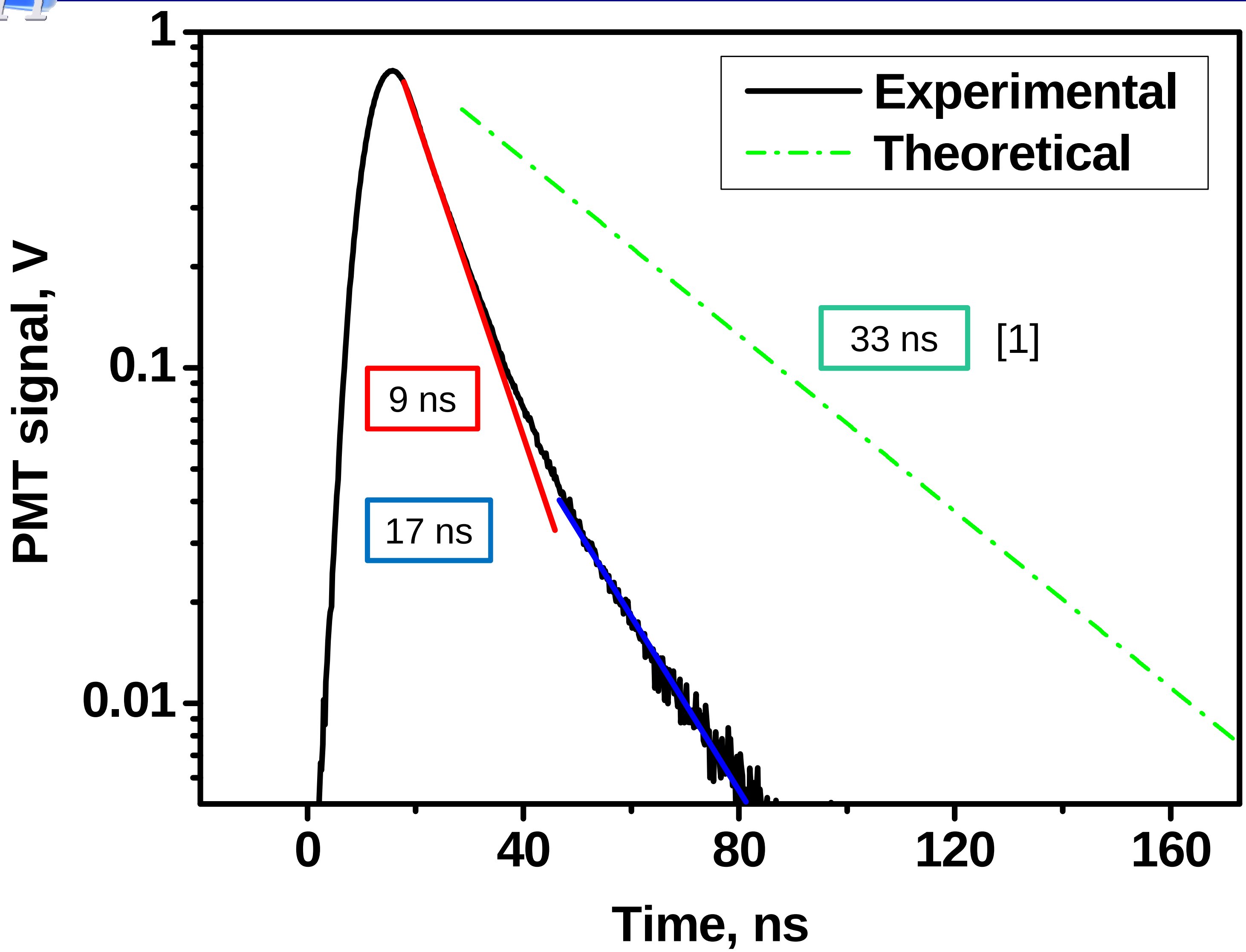


$N_2(C^3\Pi_u)$ emission decay in nitrogen



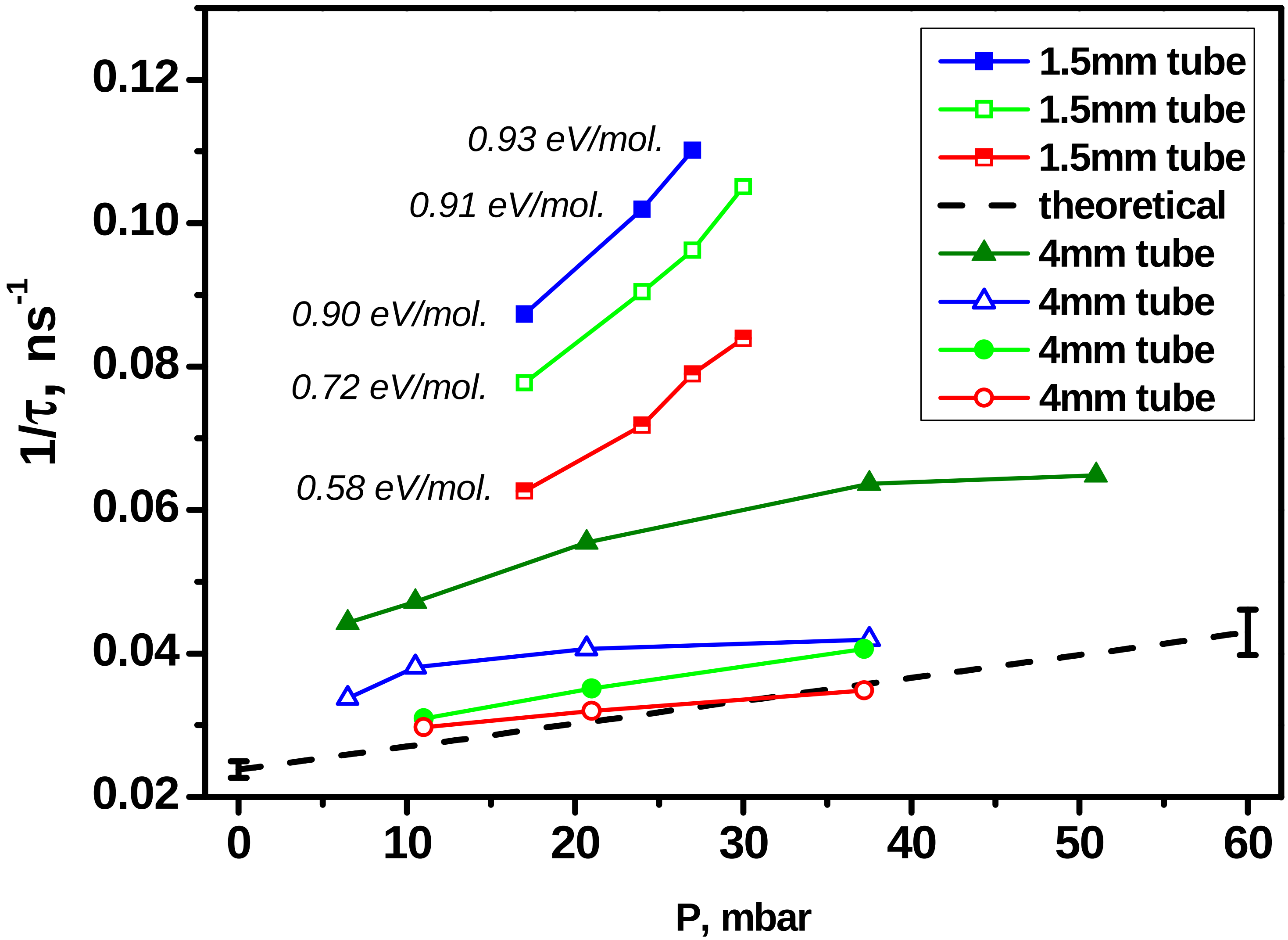
[1] Pancheshnyi, S. V., Starikovskaia, S. M., and Starikovskii, A. Y., *Chemical Physics*, 262 (2000)

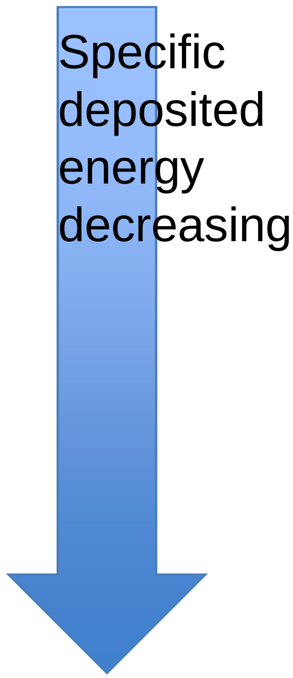
$N_2(C^3\Pi_u)$ emission decay in nitrogen



[1] Pancheshnyi, S. V., Starikovskaia, S. M., and Starikovskii, A. Yu., *Chemical Physics*, **262** (2000)

$N_2(C^3\Pi_u)$ reverse decay vs pressure





Analysis of the experimental results

Possible causes:

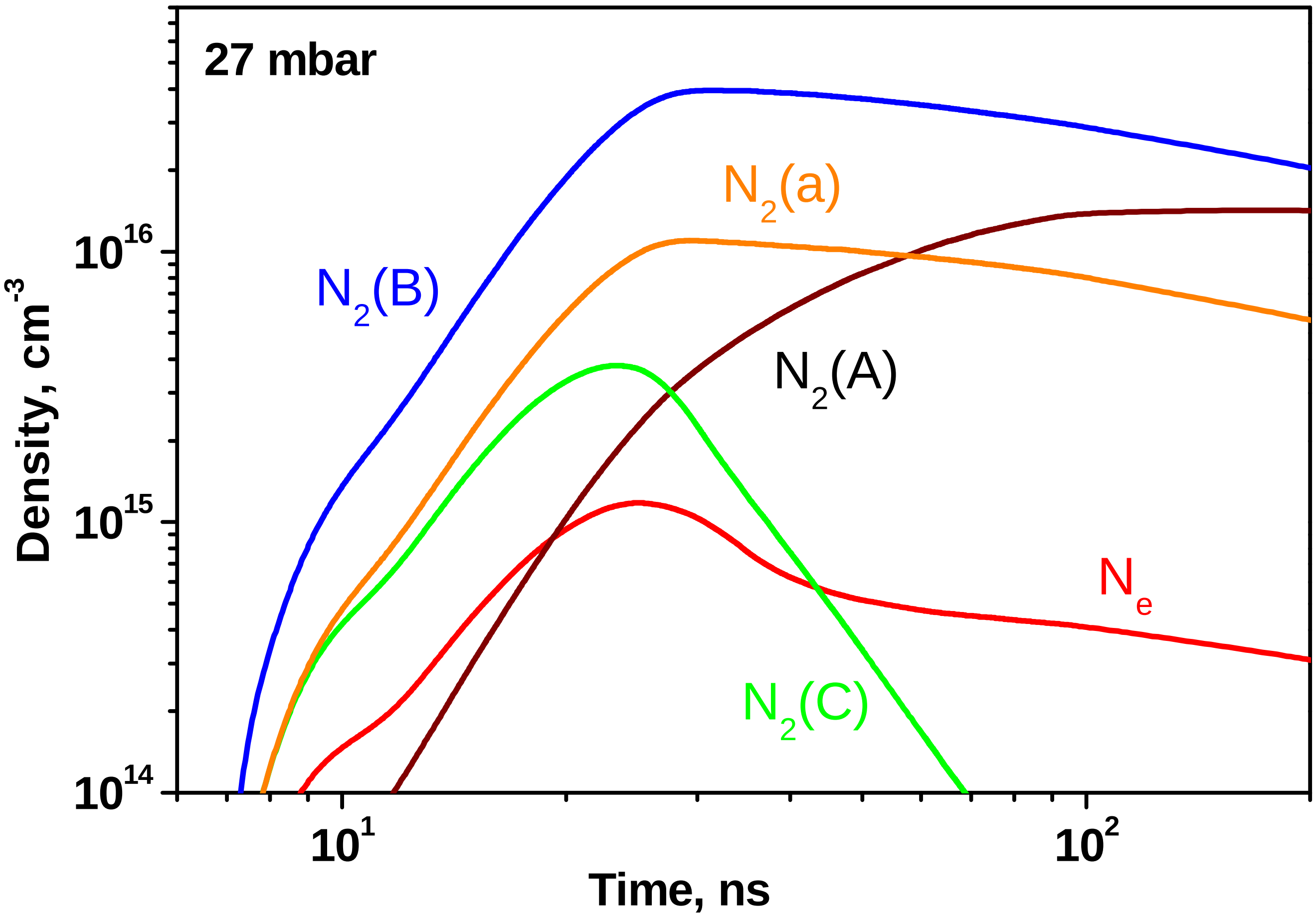
- Stimulated emission
- Variation of quenching rate with temperature
- Quenching by atoms
- Quenching by excited species (up to 10% of N_2 are electronically excited)

- Quenching by electrons: $n_e = 10^{15}$ cm^{-3}
- The electron density at t>10 ns is sustained by reactions of associative ionization:

$$N_2(A^3\Sigma^+_u) + N_2(a'^1\Sigma^-_u) \rightarrow N_4^+ + e$$

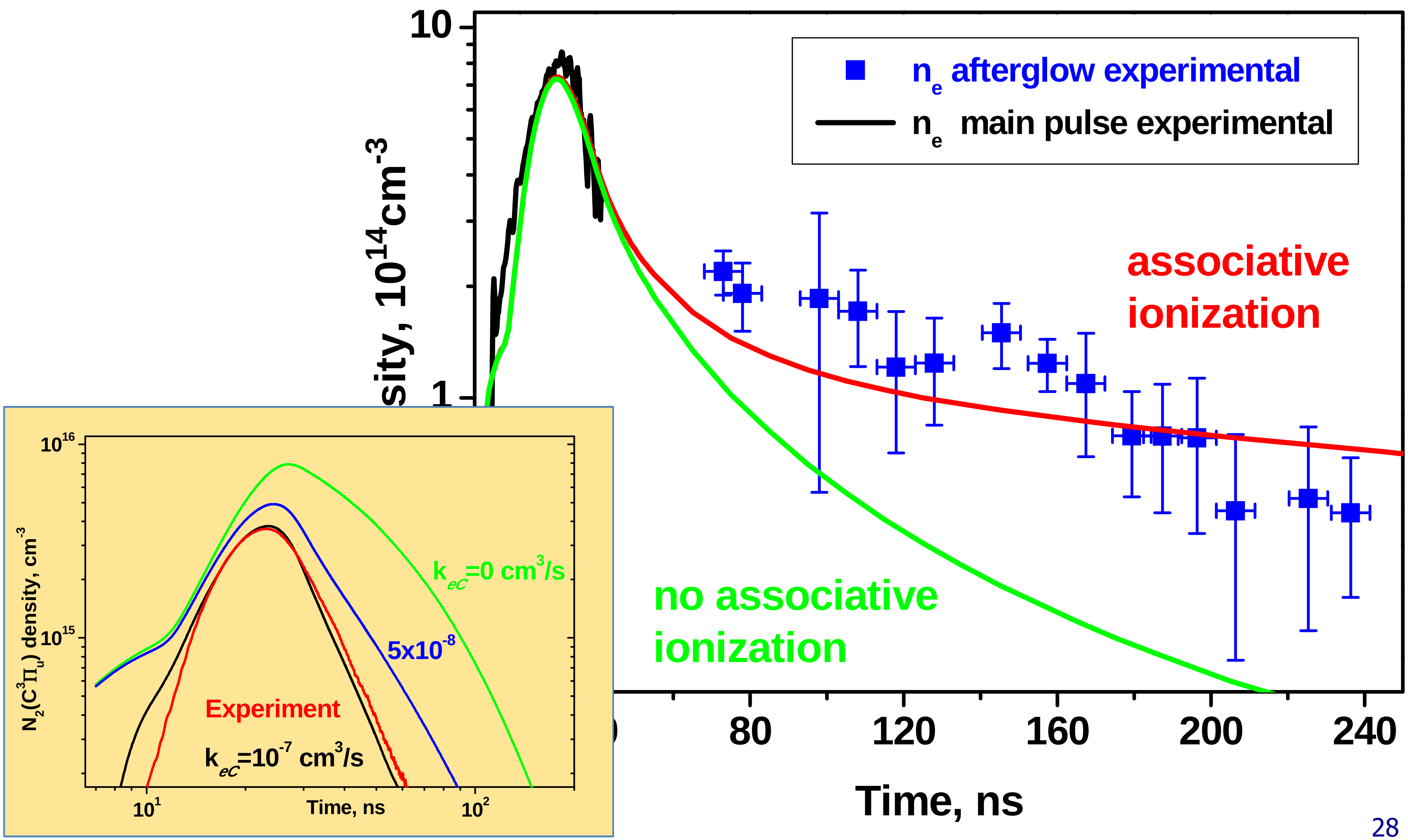
$$N_2(a'^1\Sigma^-_u) + N_2(a'^1\Sigma^-_u) \rightarrow N_4^+ + e$$

Calculated n_e and excited species densities



- electron density slightly changes after discharge pulse

Calculated and measured electron density in the afterglow in nitrogen



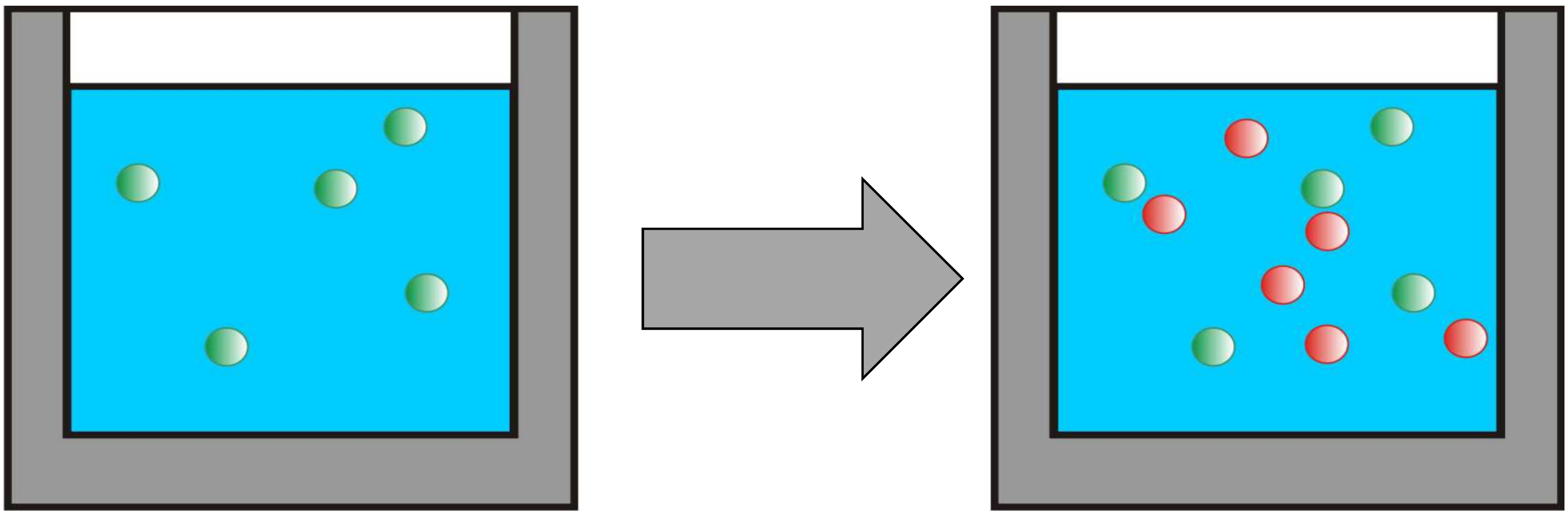
"Non-diluted" plasma chemistry: how this will influence the diagnostics?

$N_2(A^3\Sigma^+_u) + N_2(a'^1\Sigma^-_u) \rightarrow$ Prod

$N_2(C^3\Pi_u) + e \rightarrow$ Prod

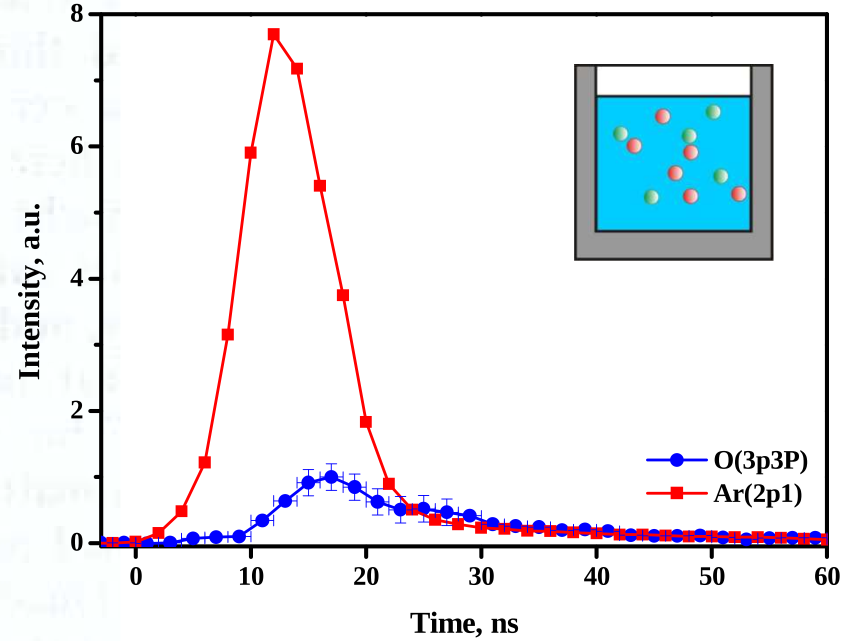
$A^* + e \rightarrow$ Prod?

$A^* + M^* \rightarrow$ Prod?



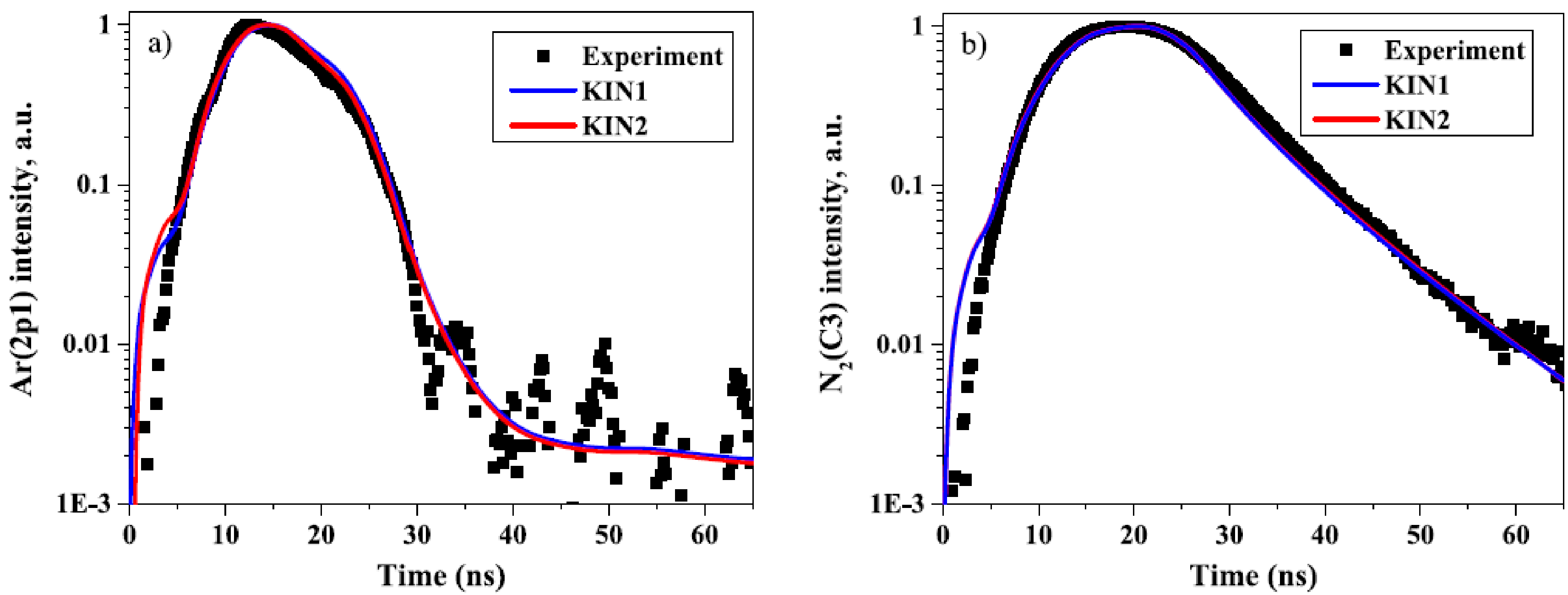
Actinometry in nanosecond capillary discharge in air, 20 Torr

$$\frac{I_{O(3p^3P)}}{I_{Ar(2p_1)}} = \alpha \frac{[O]}{[Ar]}$$



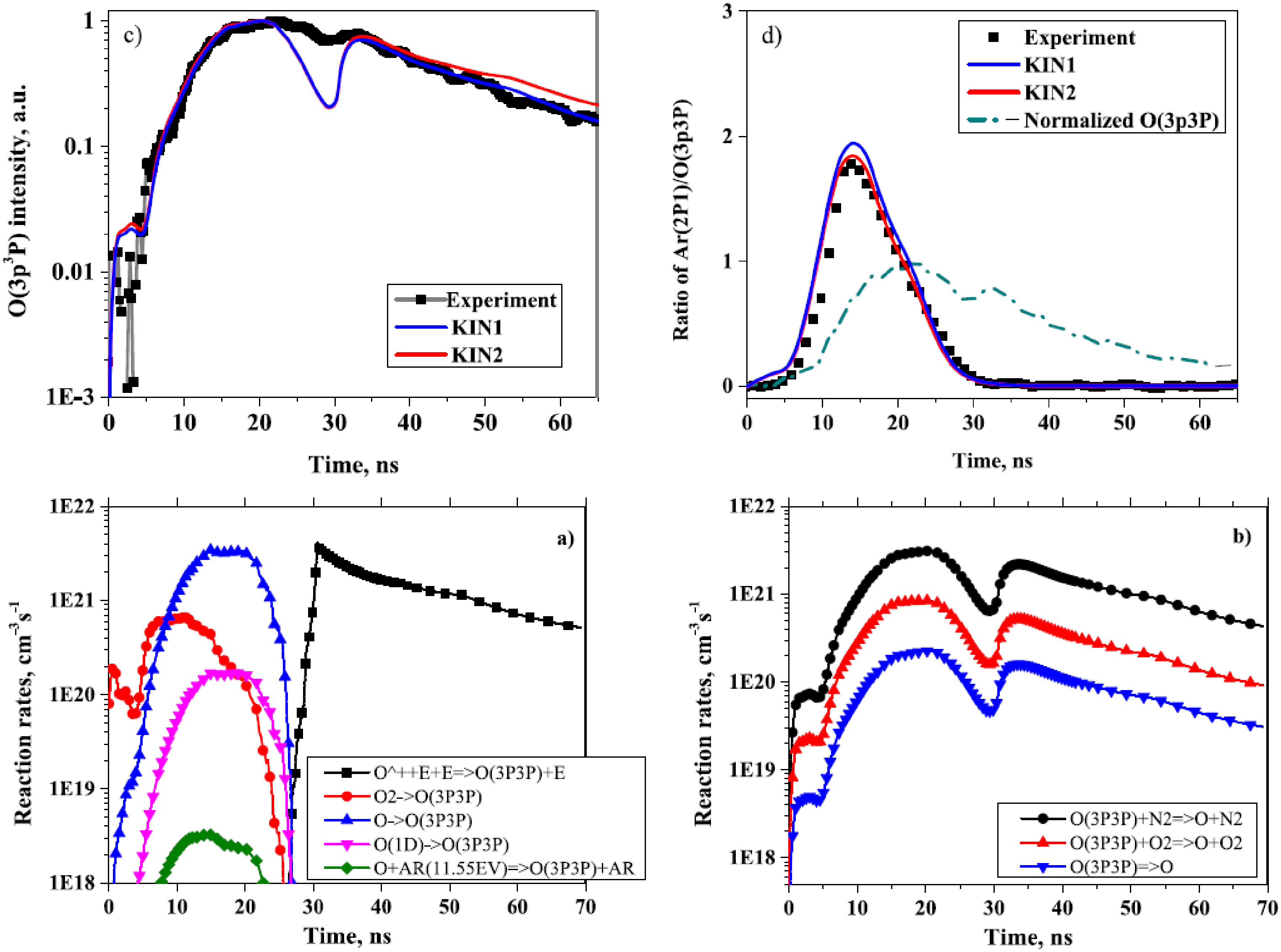
Actinometry: 5.3% Ar in air; OES of $Ar(2p_1)$, 750 nm // $O(3p^3P)$, 845 nm

Temporal behavior of Ar* and N_2(C)

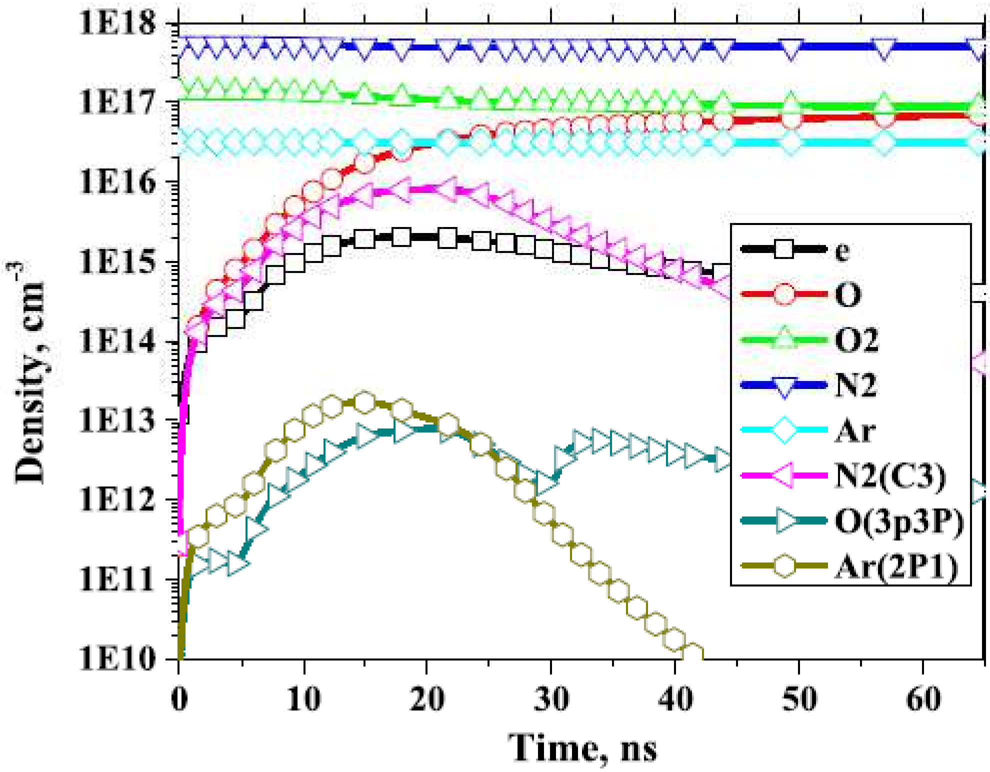


N_2, $N_2(v=1-8)$, $N_2(A^3\Sigma_u^+)$, $N_2(B^3\Pi_g)$, $N_2(a'^1\Sigma_u^-)$, $N_2(C^3\Pi_u)$, N, $N(^2D)$, $N(^2P)$, N^+, N_2^+, N_3^+, N_4^+, O_2, $O_2(v=1-4)$, $O_2(a^1\Delta_g)$, $O_2(b^1\Sigma_g^+)$, $O_2(A^3\Sigma_u^+)$, $O_2(C^3\Delta_u)$, $O_2(c^1\Sigma_u^-)$, O, $O(^1D)$, $O(^1S)$, O_3, O^+, O_2^+, O_4^+, O^-, O_2^-, O_3^-, O_4^-, NO, NO^+, NO^-, $O_2^+N_2$, N_2O, NO_2, NO_3, N_2O_5 , N_2O^+, NO_2^+, N_2O^-, NO_2^-, NO_3^-, e.

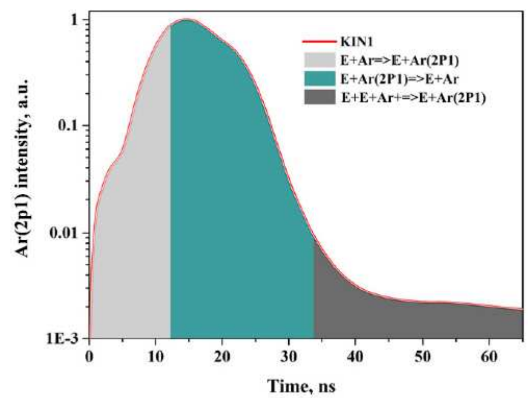
Temporal behavior of O*, Ar*/O* ratio and rates of O* production/decay



O-density: 3e16 cm-3; am expression for the discharge phase

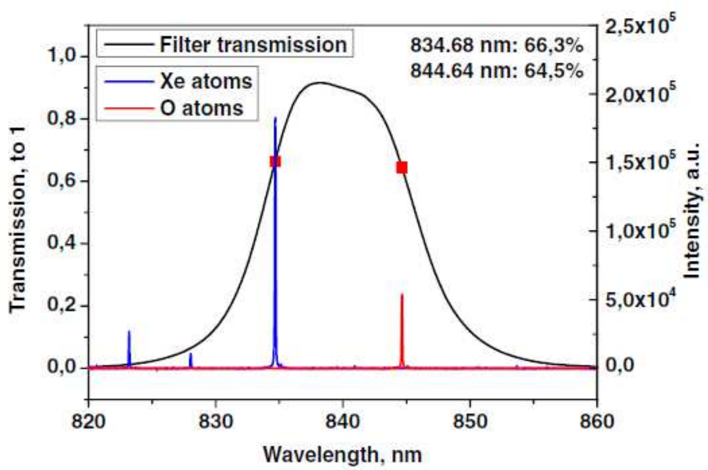


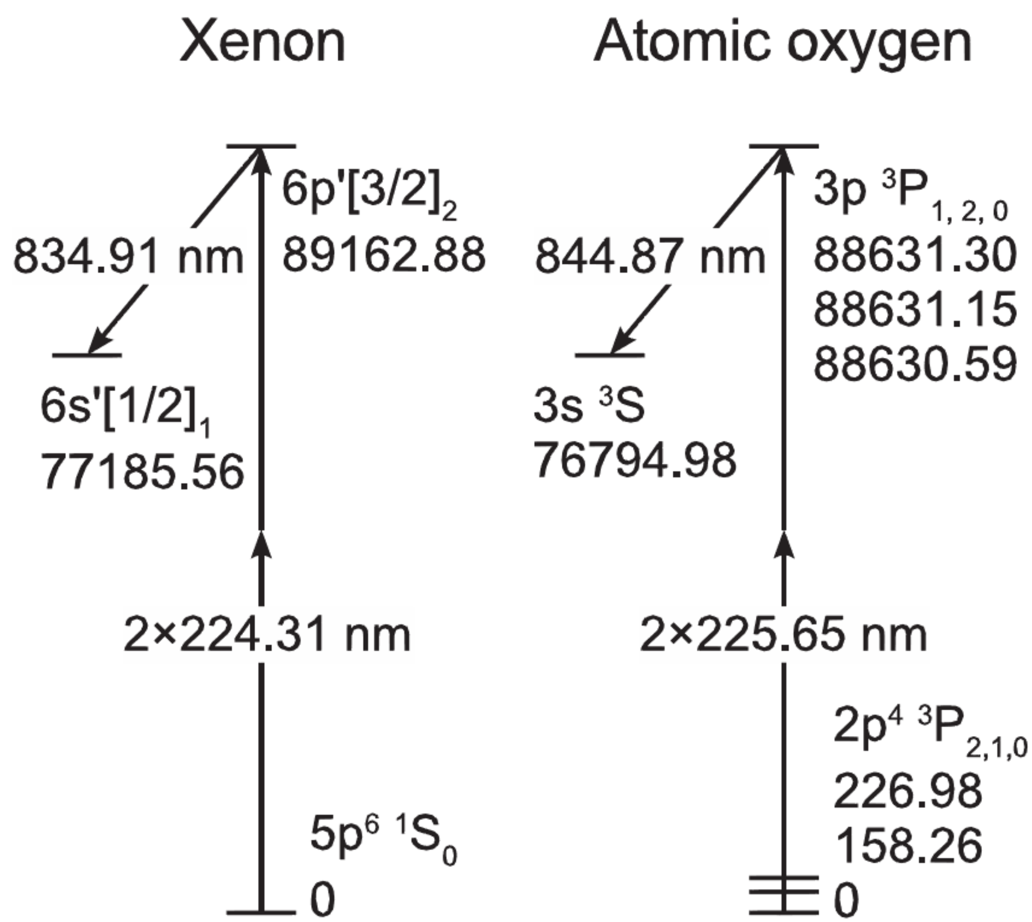
R16	$e + Ar \rightarrow e + Ar(2p_1)$
R28	$e + O \rightarrow e + O(3p^3P)$
R47	$O(3p^3P) + N_2 \rightarrow O + N_2$
R37	$Ar(2p_1) + e \rightarrow Ar + e$



$$[O] = [Ar] \cdot \frac{k_{16}}{k_{28}} \cdot \frac{d[O(3p^3P)]/dt + k_{47}[O(3p^3P)][N_2]}{d[Ar(2p_1)]/dt + k_{37}n_e[Ar(2p_1)]}$$

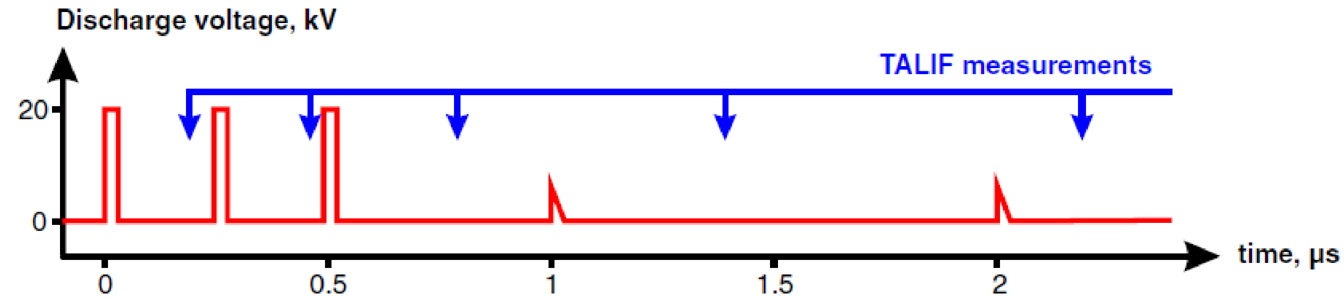
O-TALIF: scheme of the levels

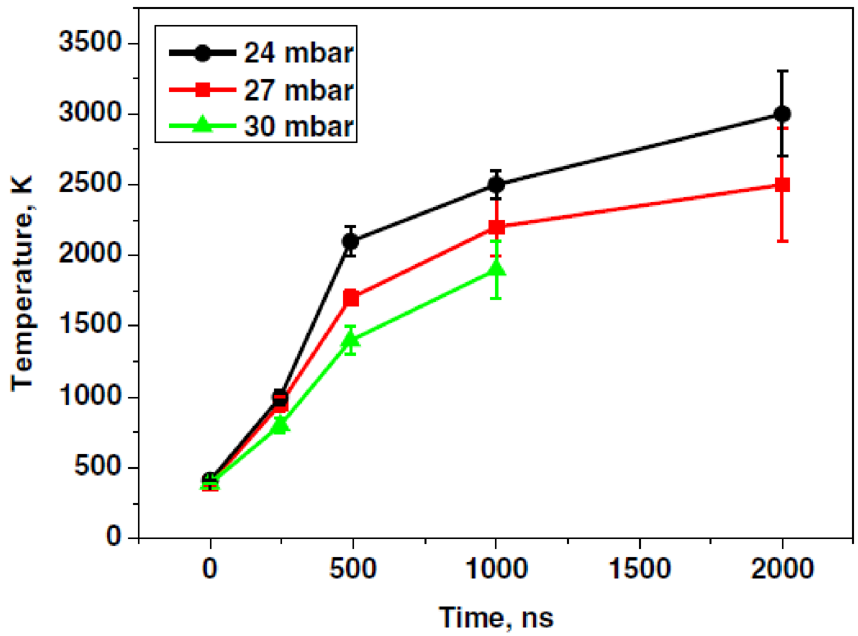


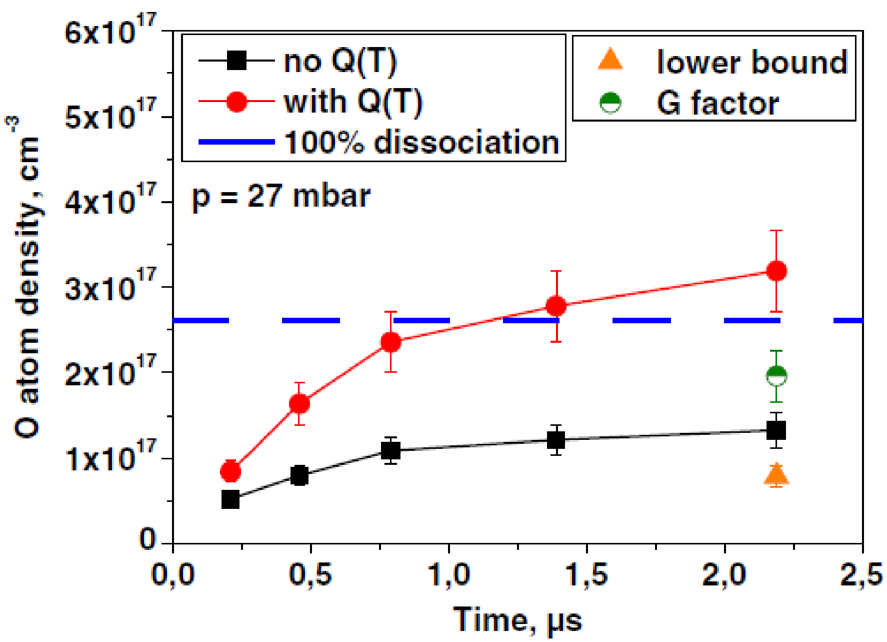


Niemi, K., Schulz-von der Gathen, V., and Döbele, H. F., *Plasma Source Science and Technology*, Vol. **14**, No. 2, 2005, pp. 375-386

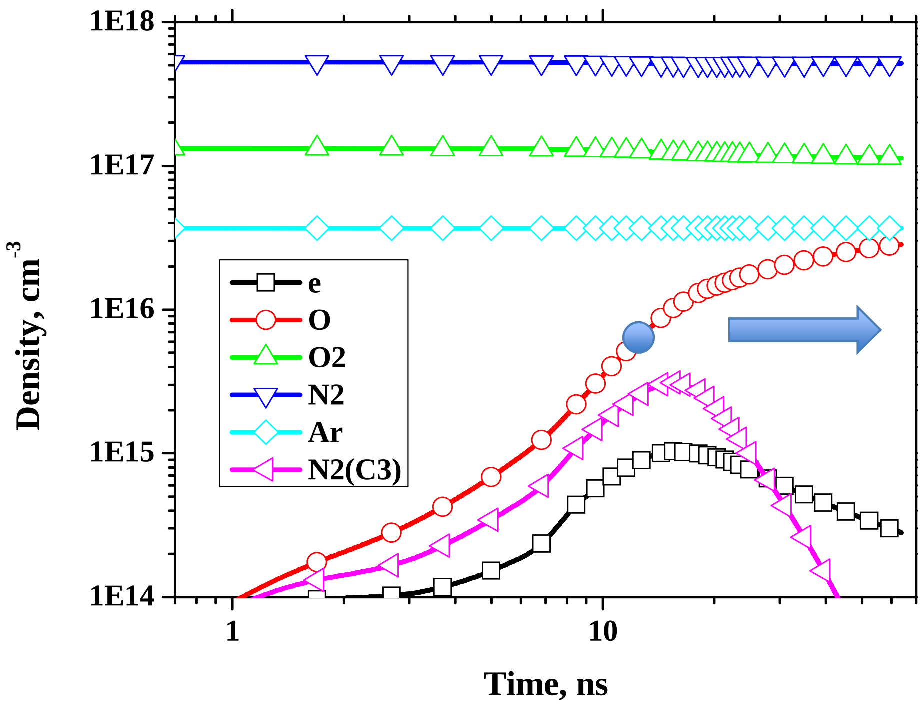
TALIF: O-atoms density in the afterglow

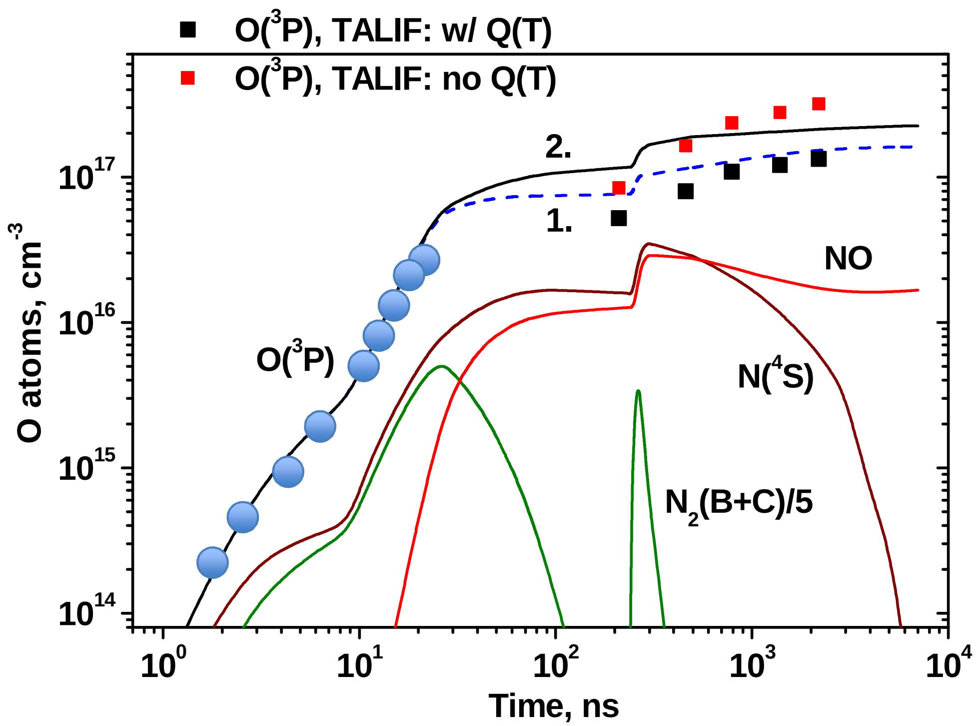






Long time trend: actinometry and TALIF





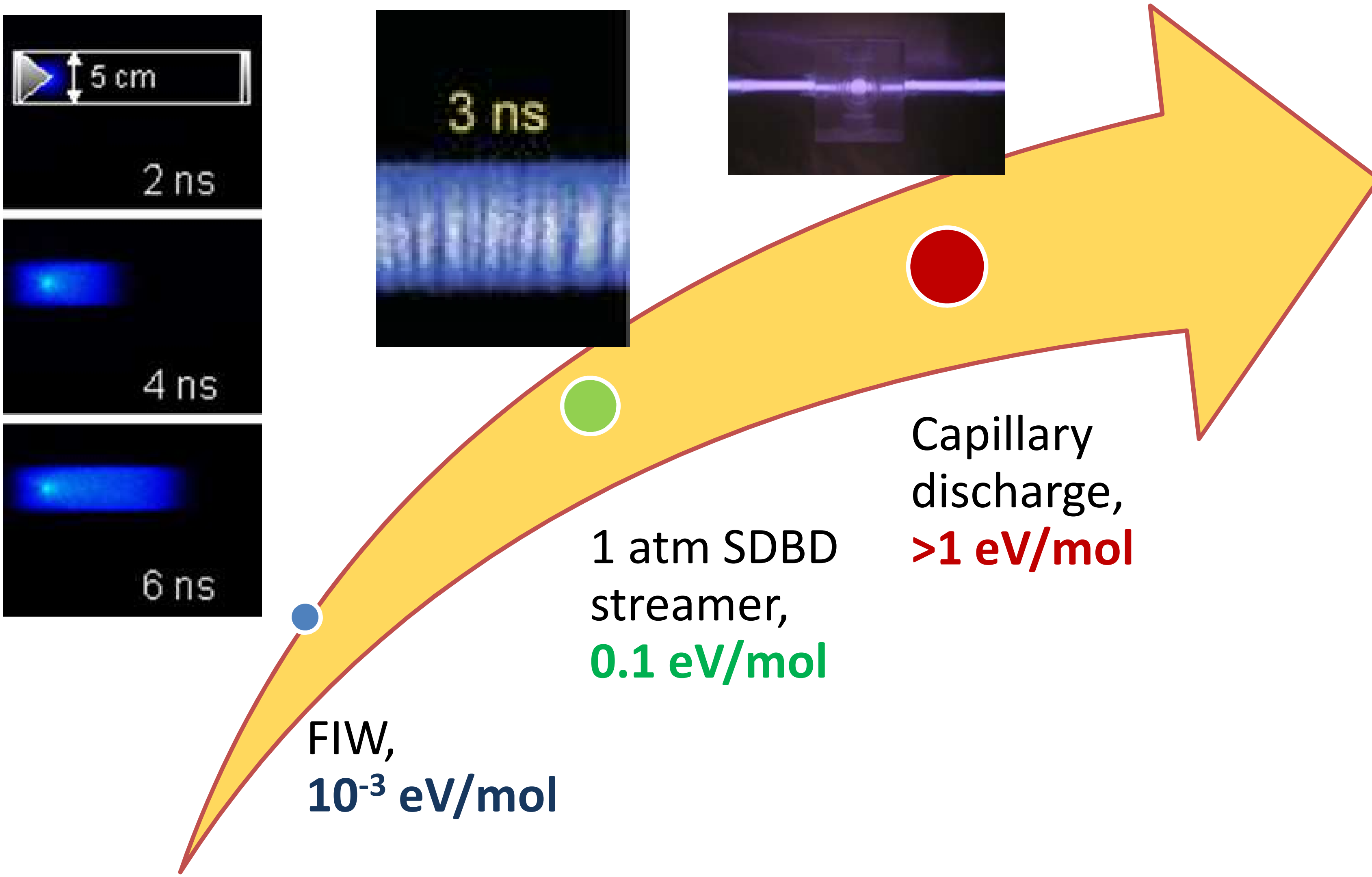
(R4)	$N_2(B^3\Pi_g) + O_2 \rightarrow N_2(v) + 2O(^3P)$	$3.0x10^{-10}$
(R5)	$N_2(a'^1\Sigma_u^-) + O_2 \rightarrow N_2(v) + O(^3P) + O(^1D)$	$2.8x10^{-11}$
(R6)	$N_2(C^3\Pi_u) + O_2 \rightarrow N_2 + O(^3P) + O(^1D)$	$2.5x10^{-10}$

Capillary discharge (U=+10 kV in the cable, T=30 ns)

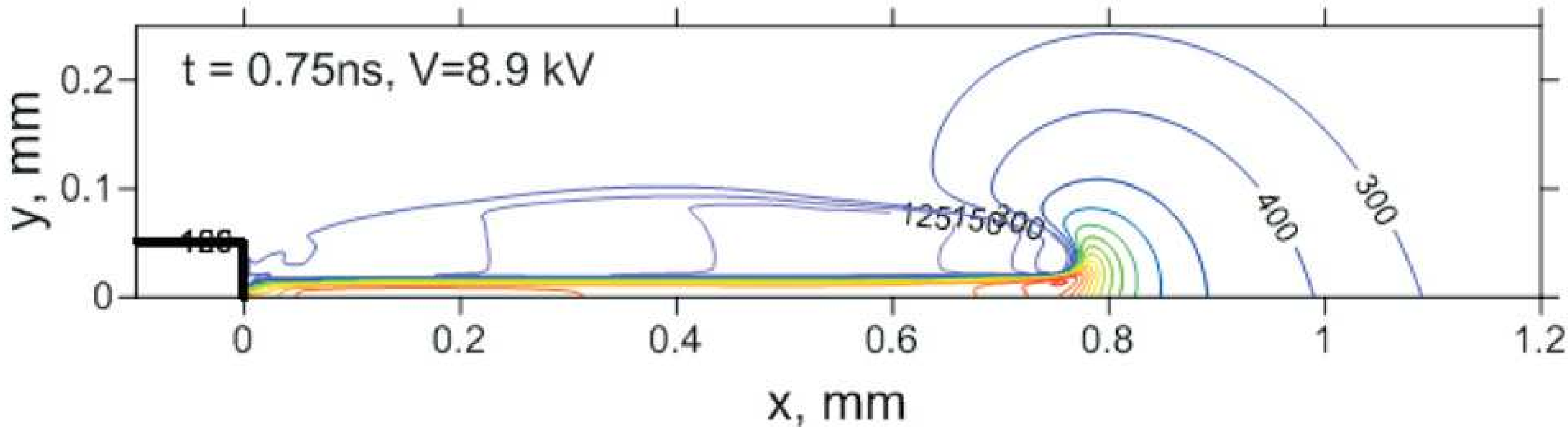
Discharge in air at 27 mbar ($N=7\times10^{17}$ cm^{-3}):

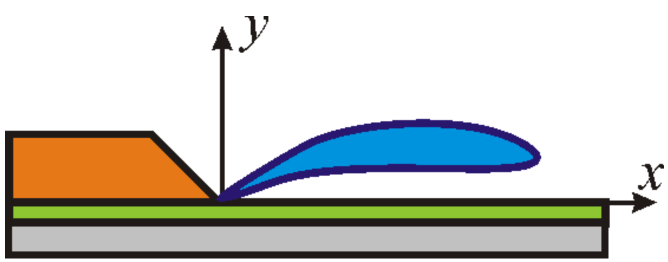
I(t): 60 A (2.5 kA/cm^2);
v(t,x): 15 mm/ns;
E/N(t,x): 1-3 kTd in the front and 200-400 Td in the region of energy deposition after the front;
$n_e(t,x)$: 10^{15} cm^{-3};
[O](t): 50-100% dissociation of O_2 at 2 μs

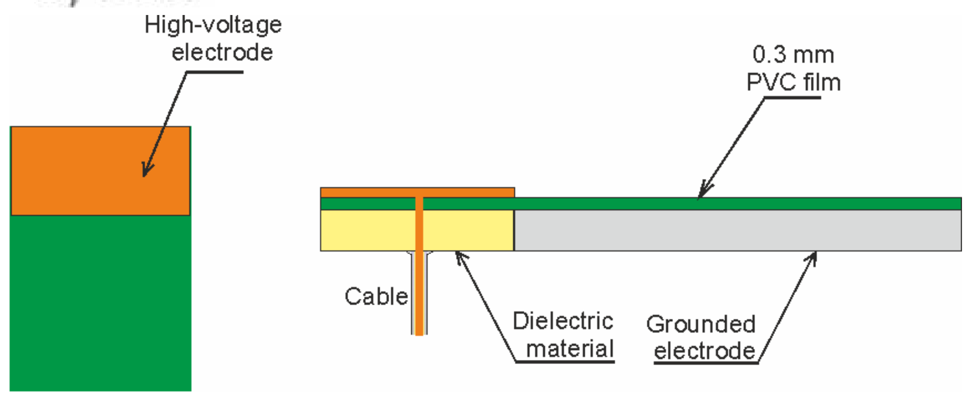
Scale of specific deposited energy



Atmospheric pressure nanosecond SDBD

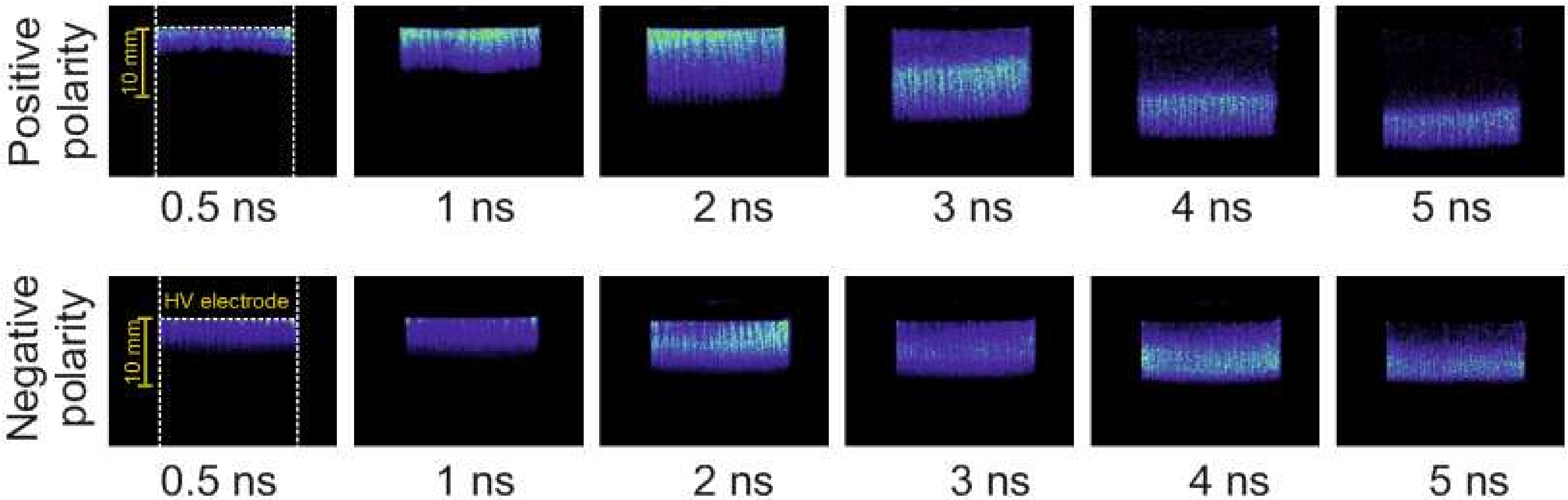


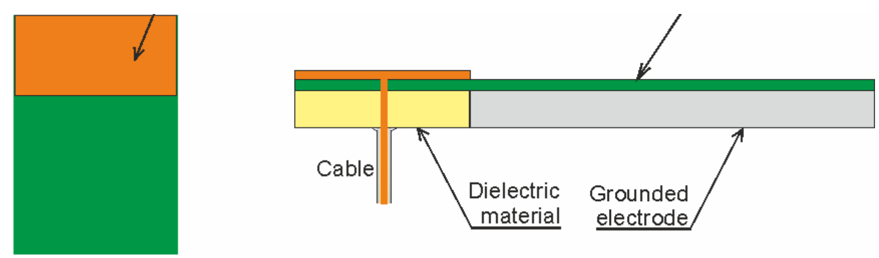




2014 S.A.Stepanyan, **V.R.Soloviev** and S.M.Starikovskaia J.Phys.D.: Appl. Phys, 47, 485201 (13pp)

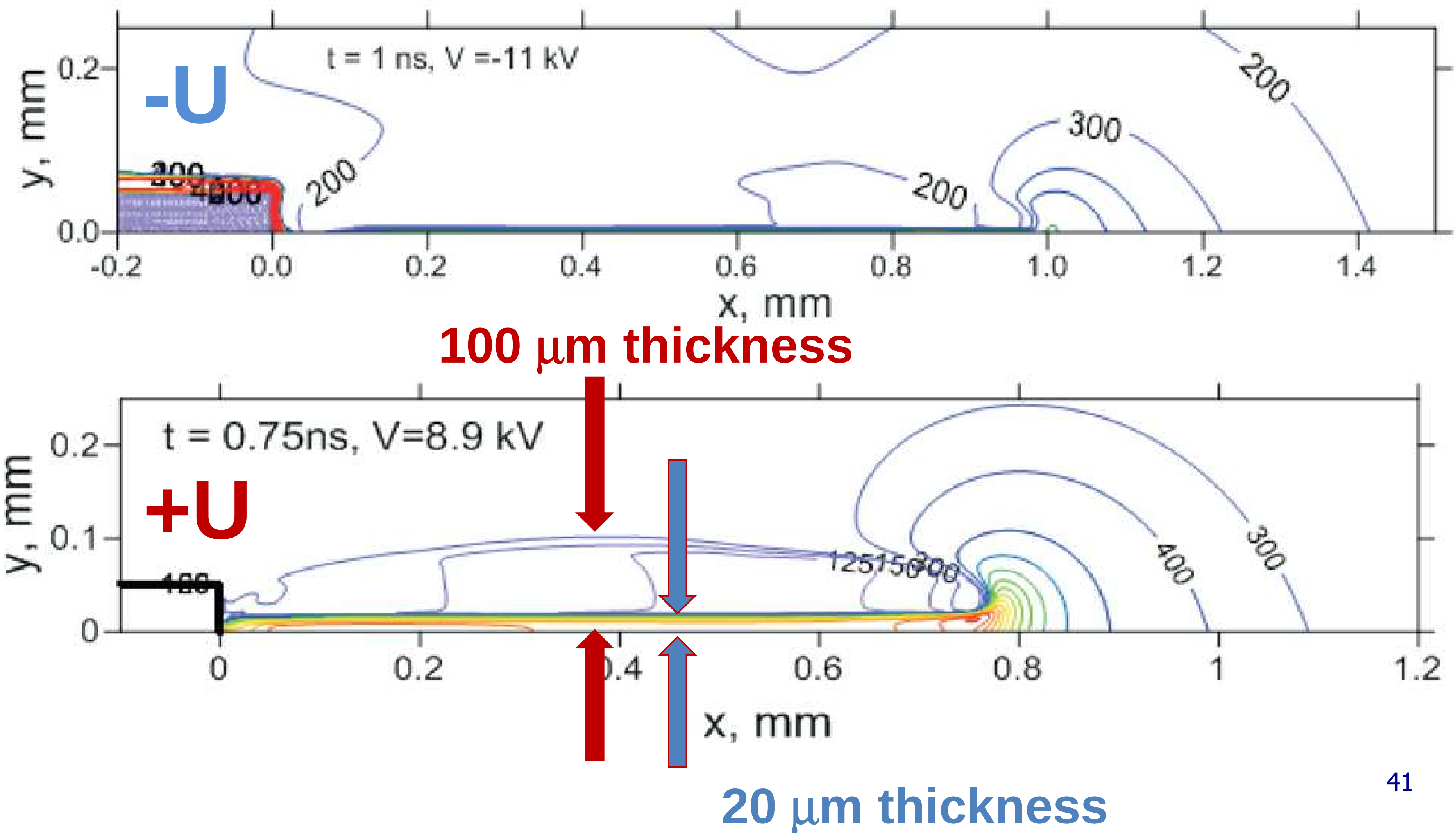
Propagation by ICCD imaging (ICCD gate is 1 ns, $N_2(C^3\Pi_u)$ emission)



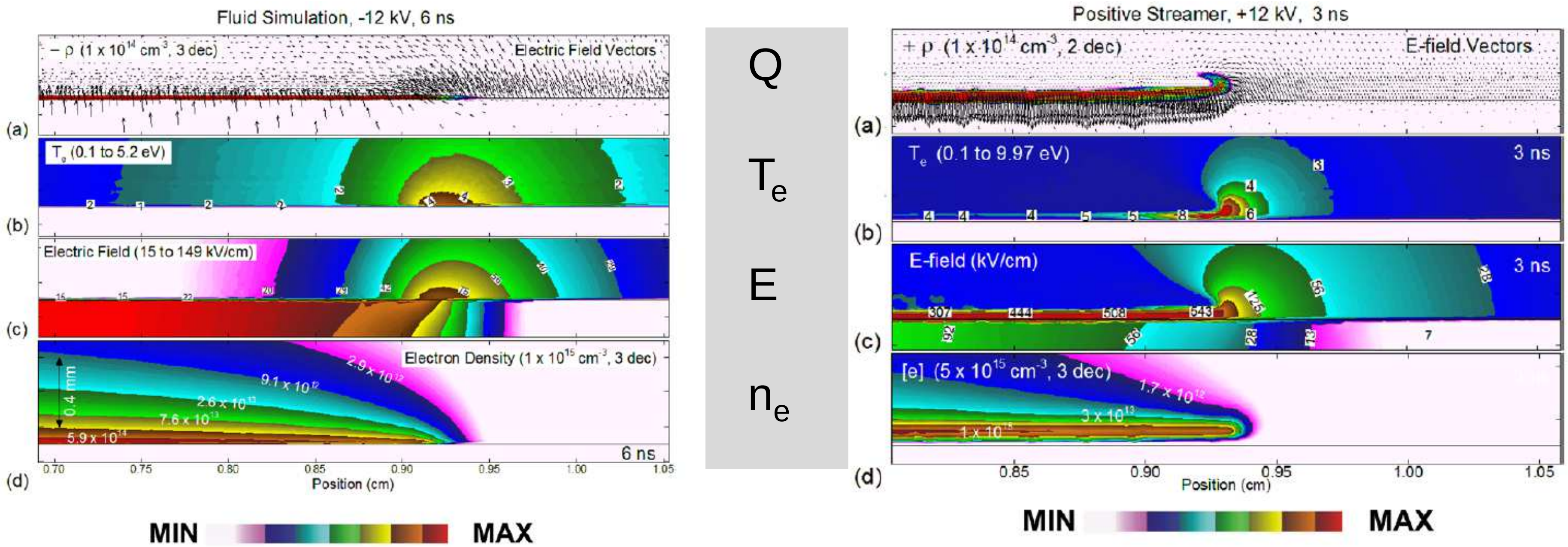


2003 First time: D.V. Roupassov, A.Yu.Starikovskii, MIPT
(GE Project //PhD Thesis)

nSDBD modeling; difference between –U and +U

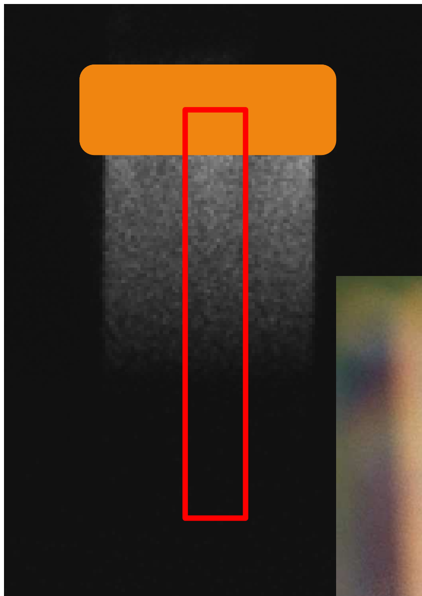


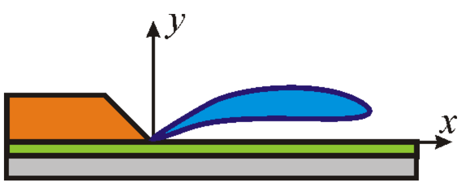
Distribution of parameters near the surface streamer head



2016 N.Yu.Babaeva, D.V.Tereshonok and G.V.Naidis
Plasma Sources Sci. Technol. 25 044008

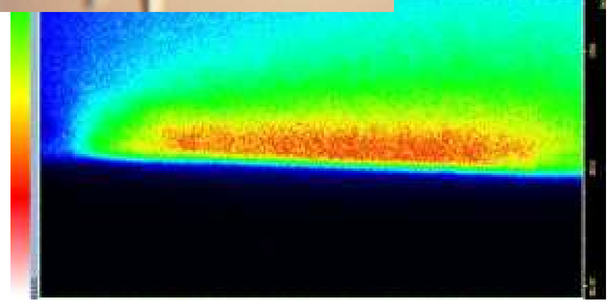
nSDBD ICCD imaging: 2^+ system of N_2



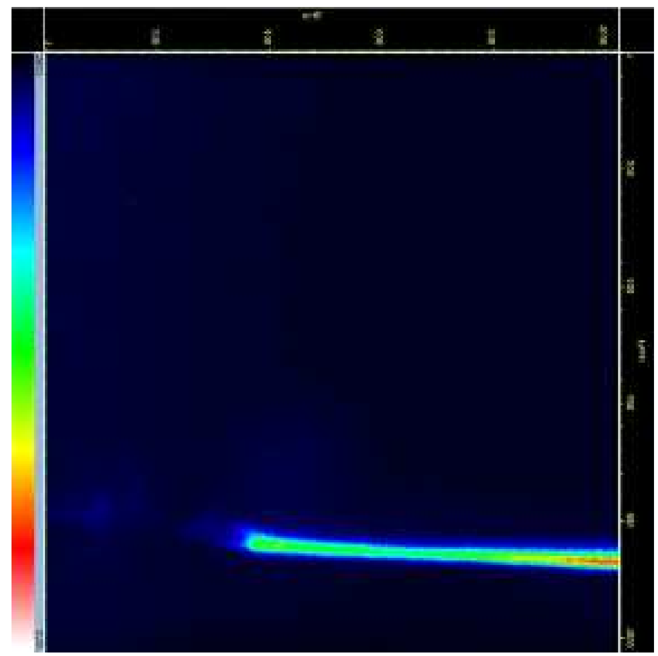


0.5 ns gate, (7x7) mm per pixel

No diaphragm

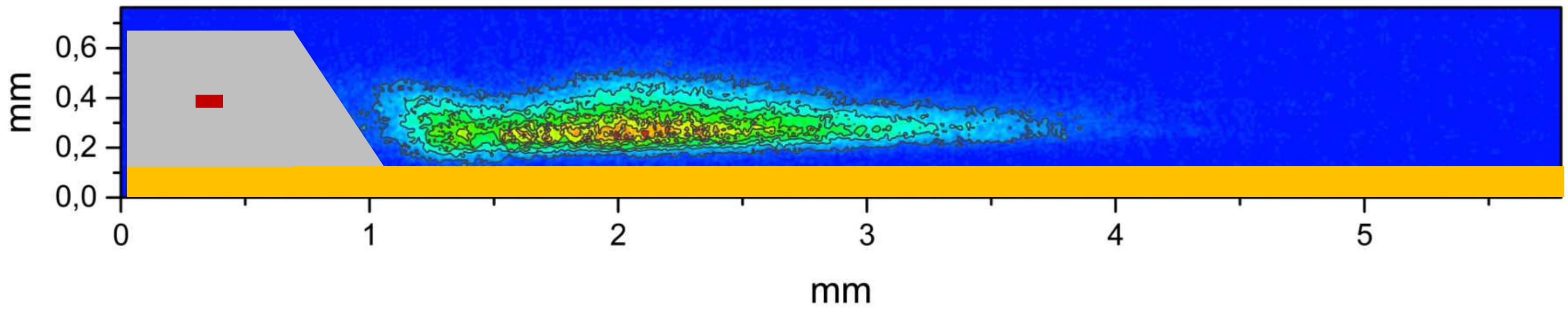


With diaphragm

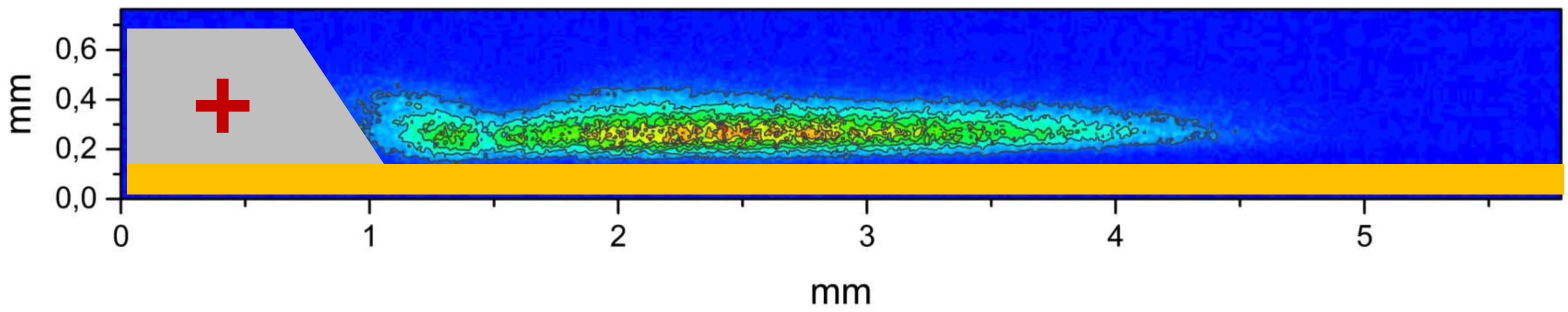


nSDBD development: 1 atm air, |U|=24 kV

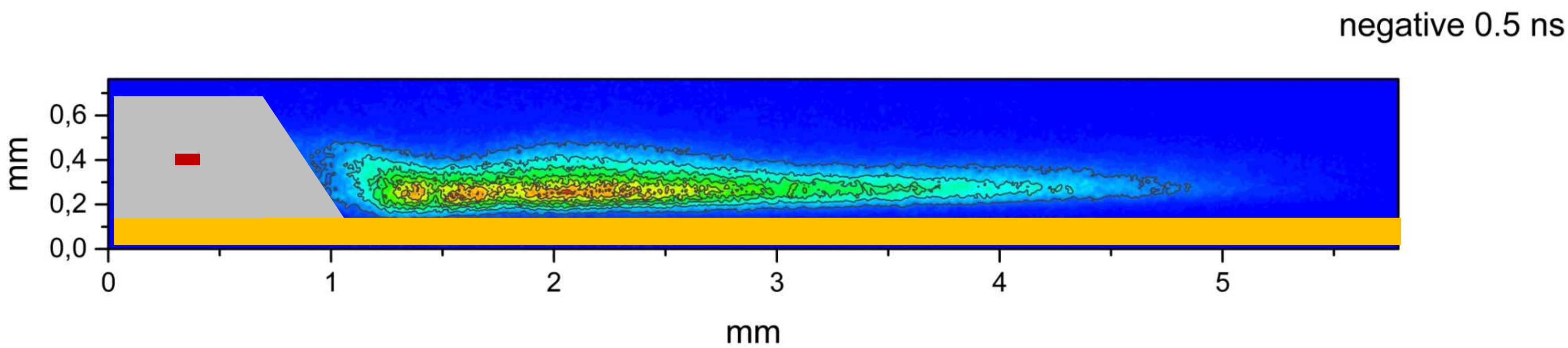
negative 0.0 ns

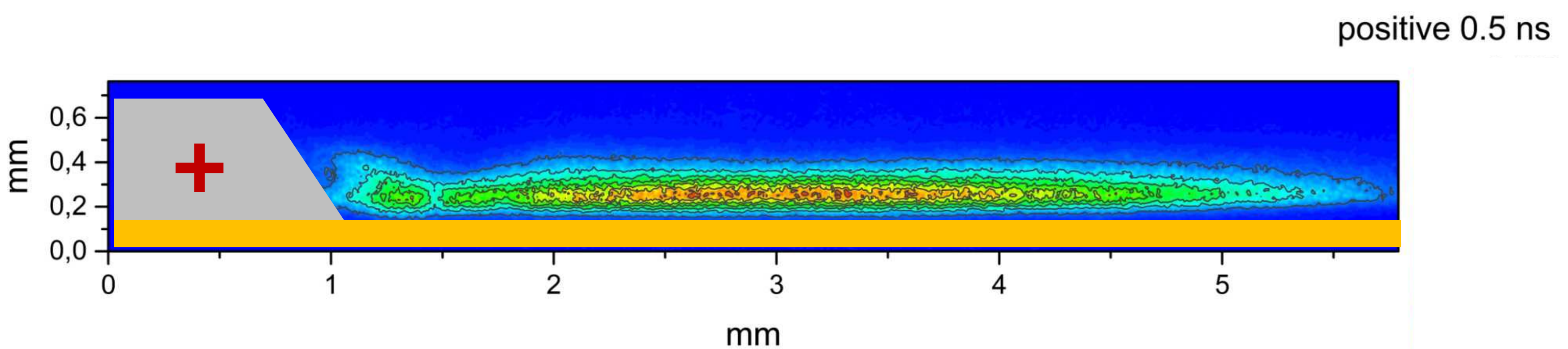


positive 0.0 ns



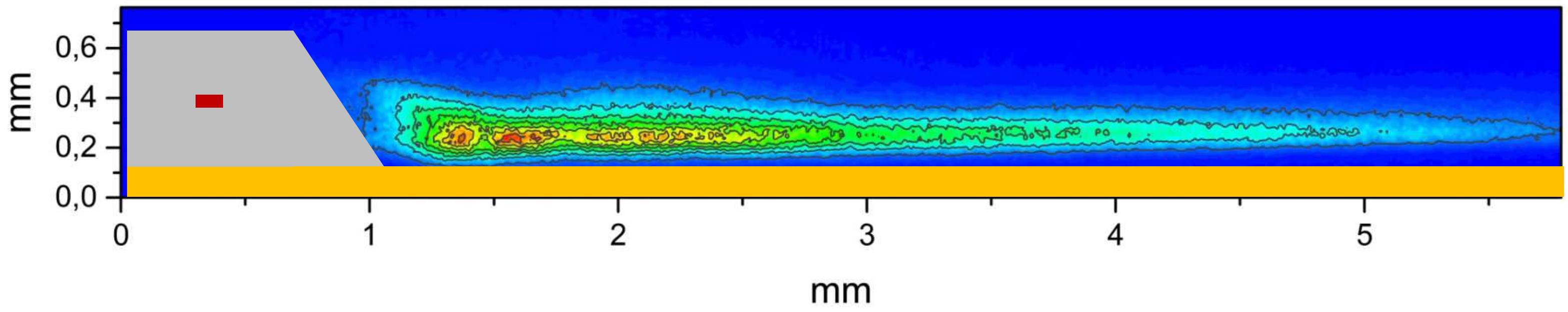
nSDBD development: 1 atm air, |U|=24 kV



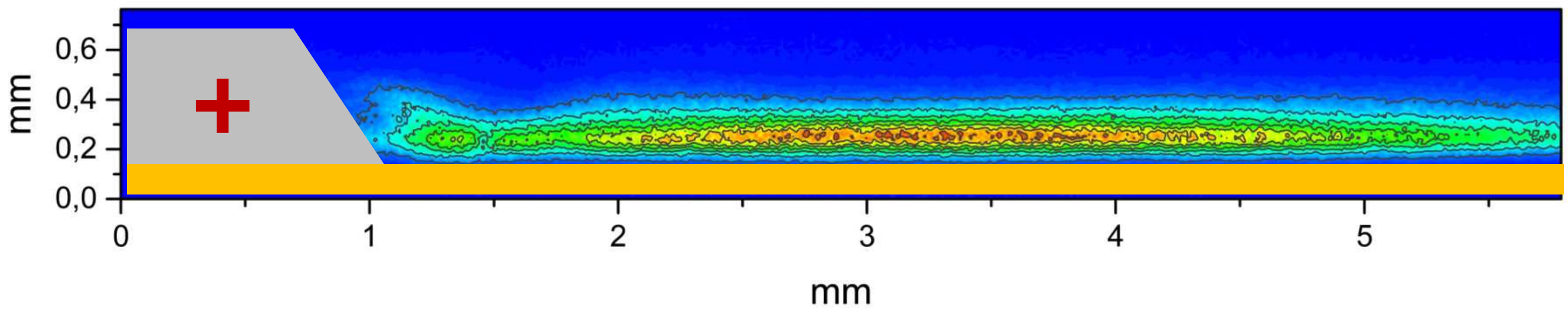


nSDBD development: 1 atm air, |U|=24 kV

negative 1.0 ns

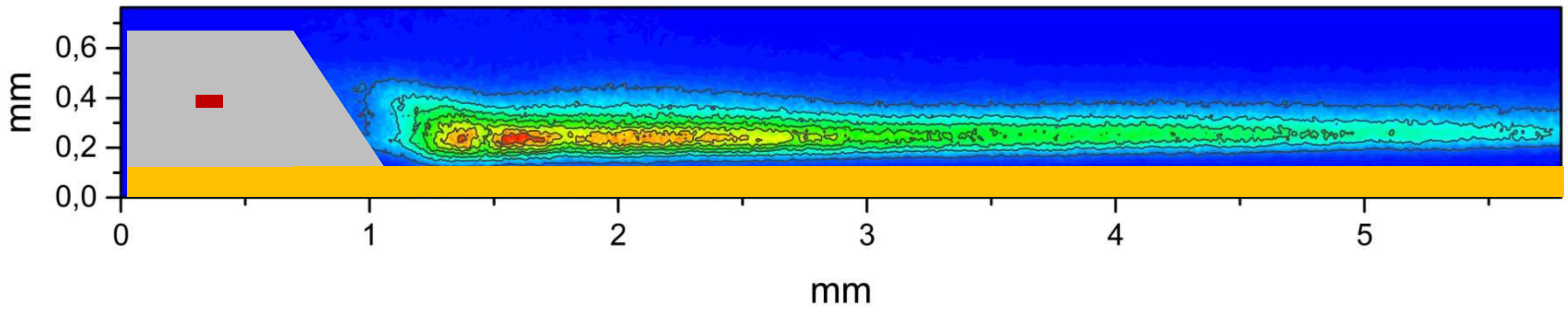


positive 1.0 ns

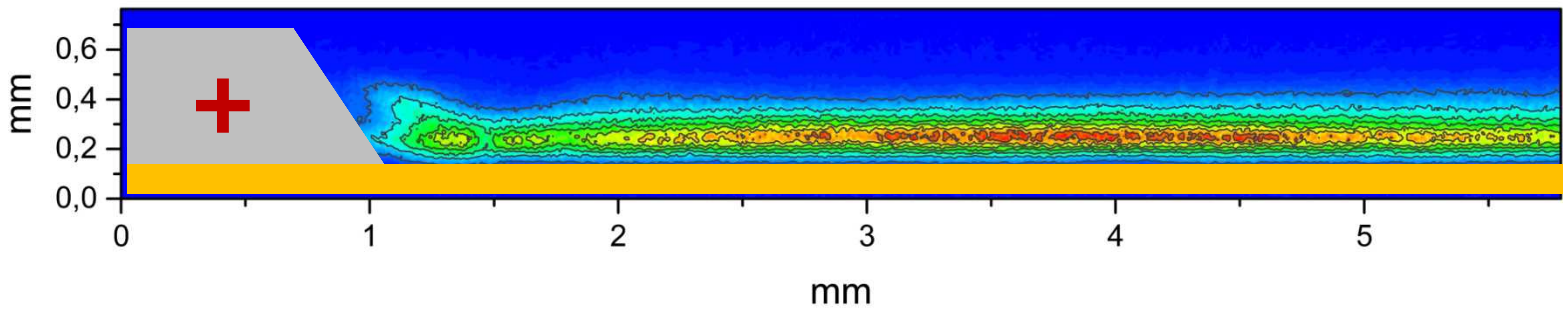


nSDBD development: 1 atm air, |U|=24 kV

negative 1.5 ns

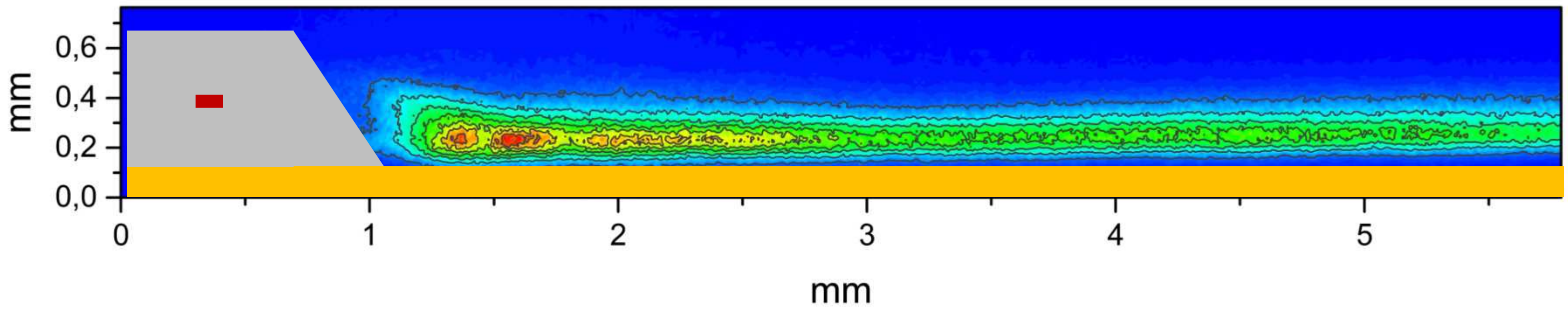


positive 1.5 ns

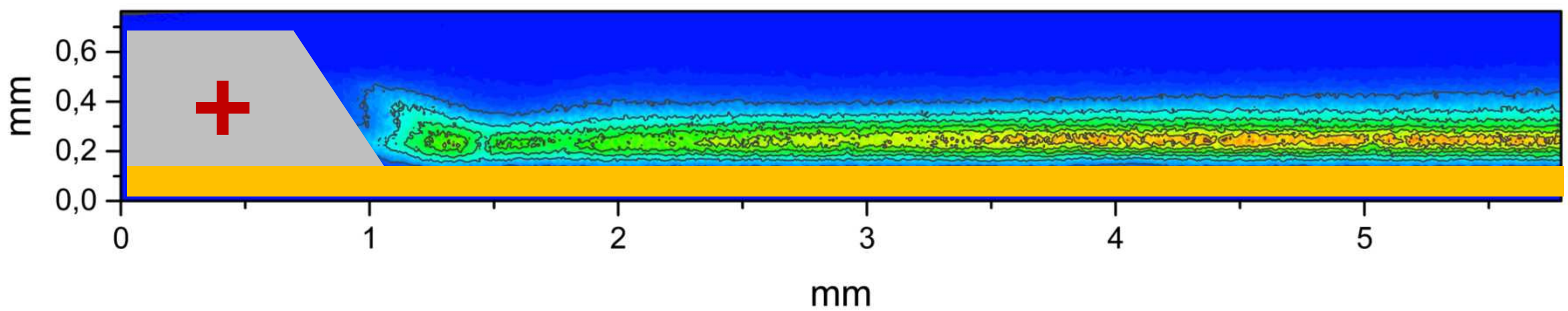


nSDBD development: 1 atm air, |U|=24 kV

negative 2.0 ns

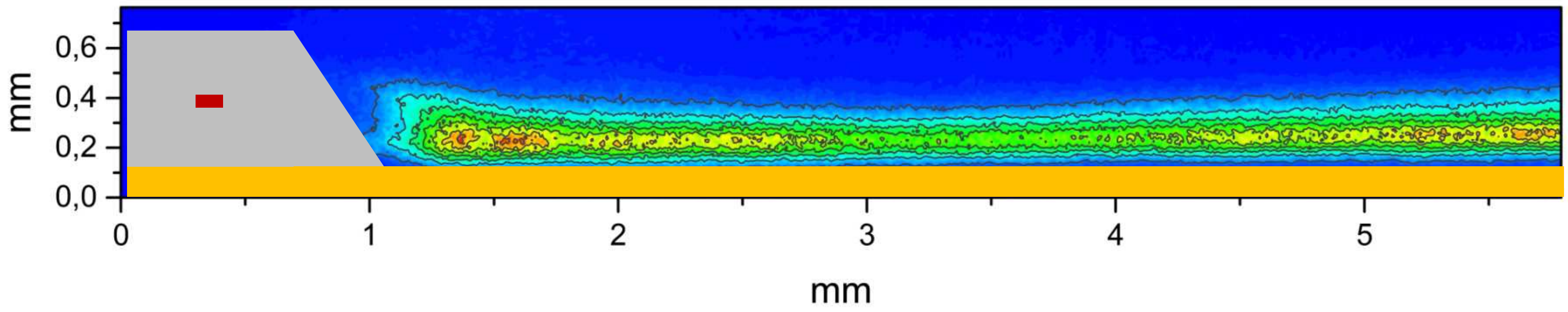


positive 2.0 ns

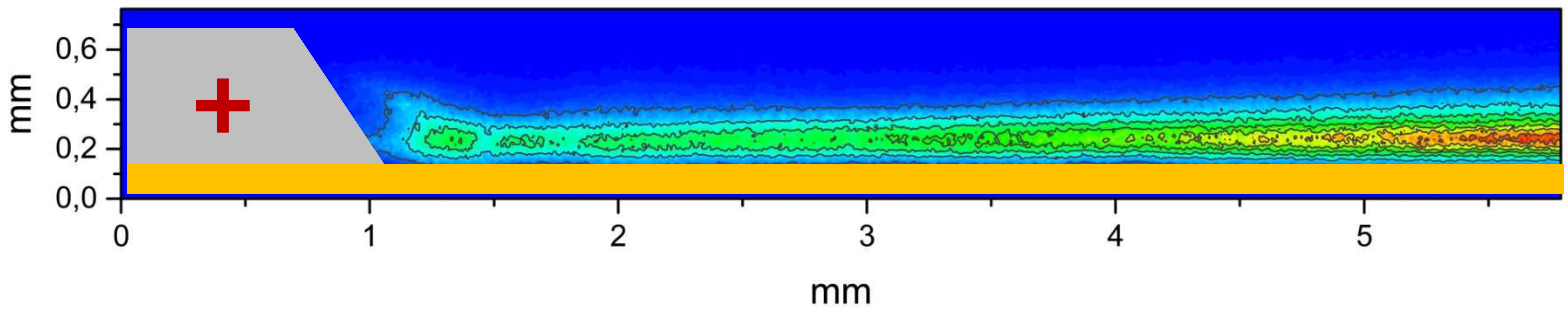


nSDBD development: 1 atm air, |U|=24 kV

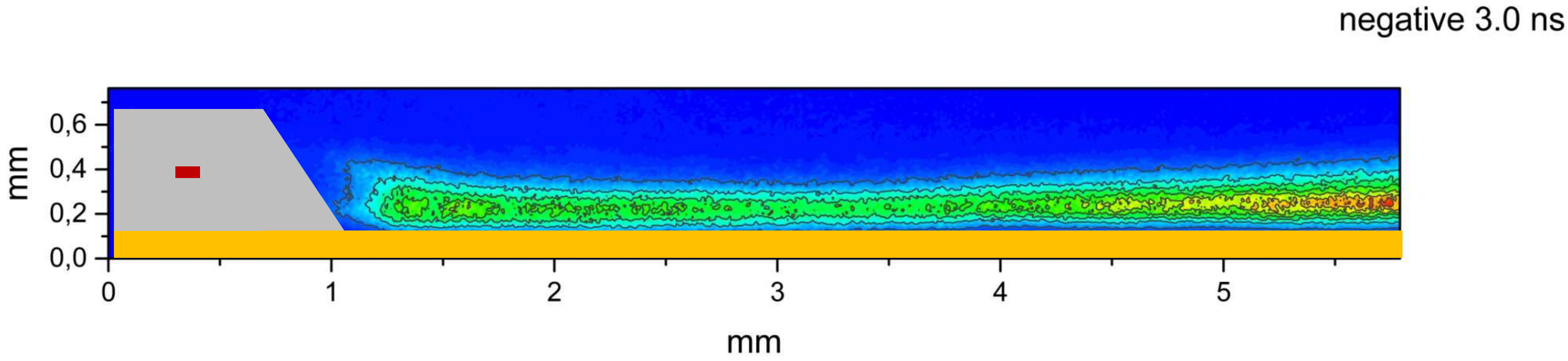
negative 2.5 ns



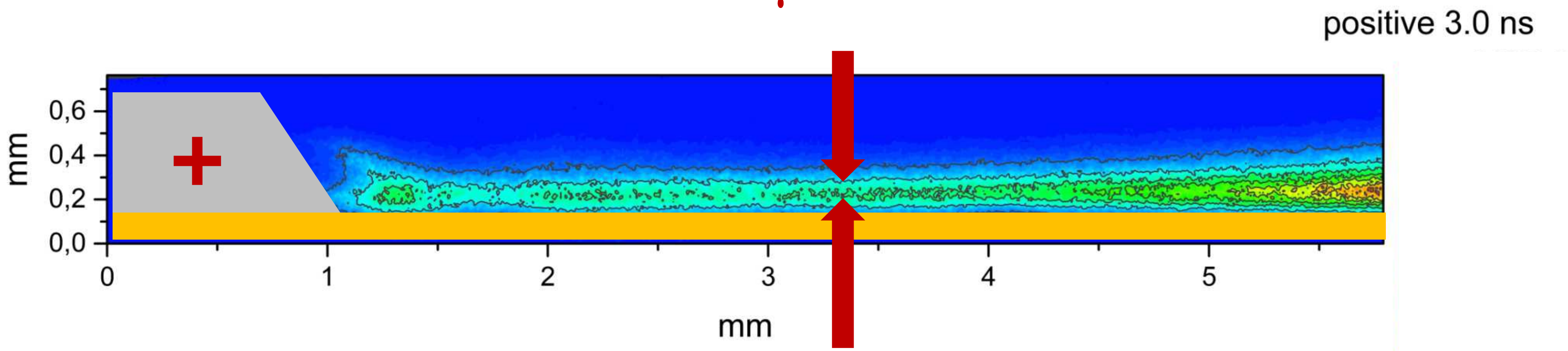
positive 2.5 ns



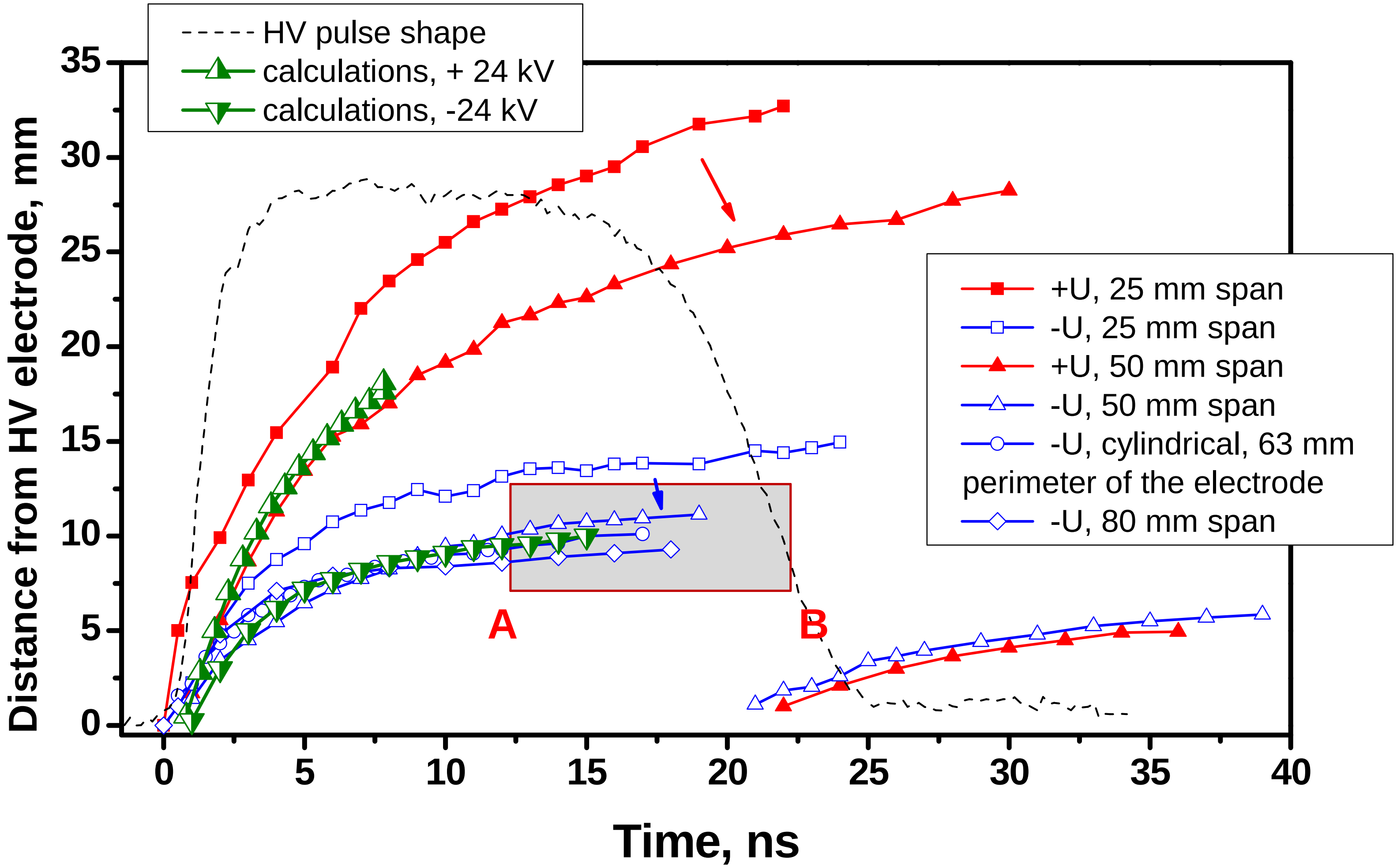
nSDBD development: 1 atm air, |U|=24 kV



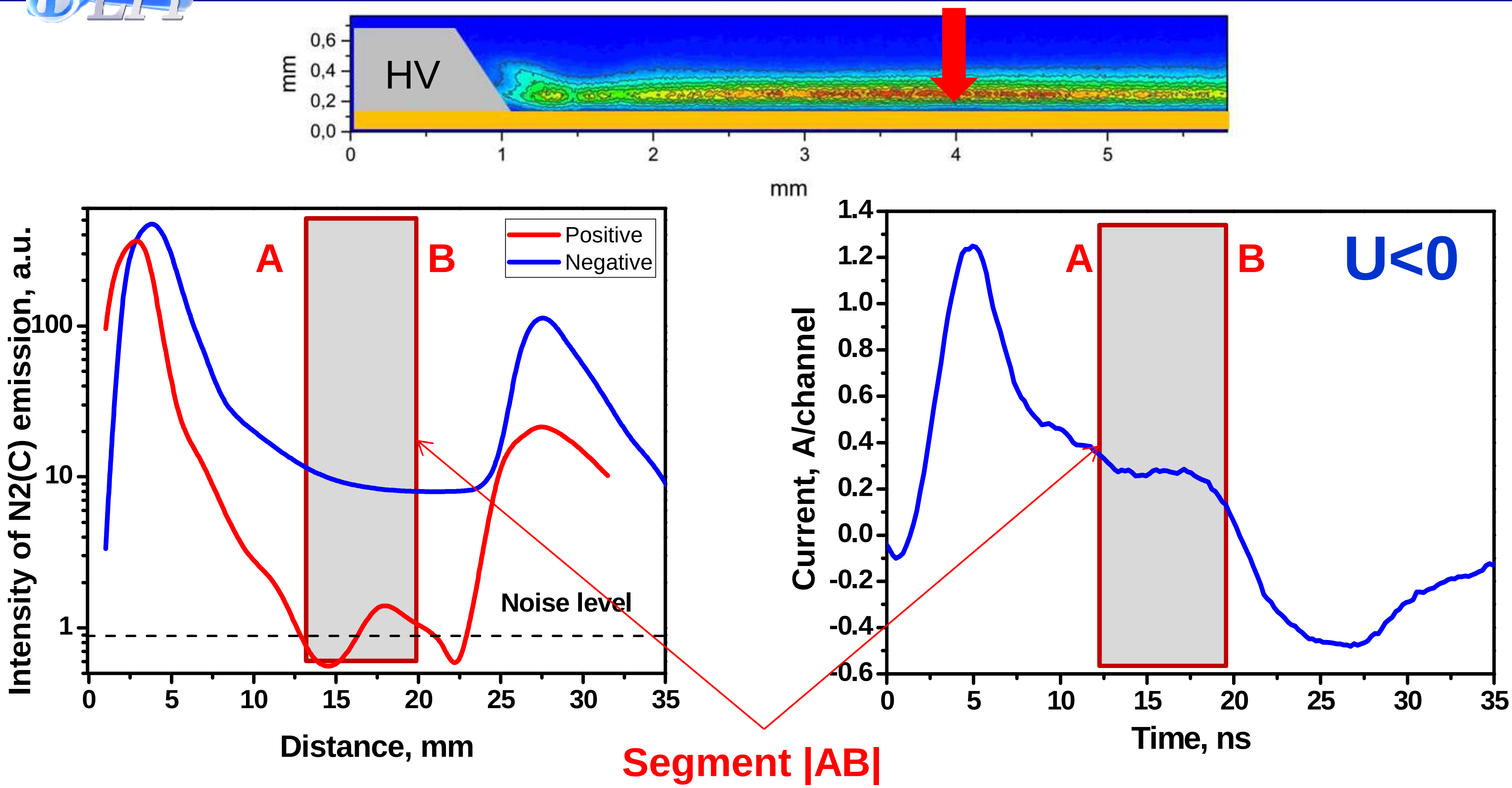
100 μm thickness



x-t diagram: streamer head *vs* time



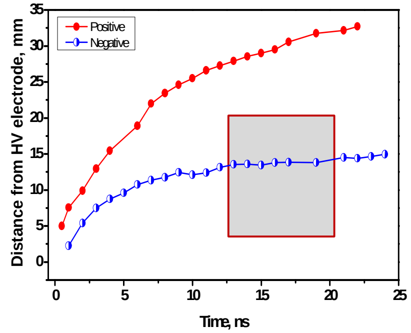
$N_2(C^3\Pi_u)$ emission 3 mm away from HV electrode and current in the streamer

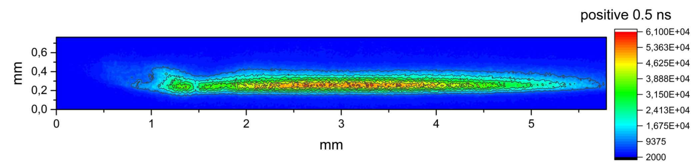


Streamer (U<0) stops after ≈10 ns.
Emission intensity and electrical current on |AB| are ≈*const*

What we can estimate from the experimental data

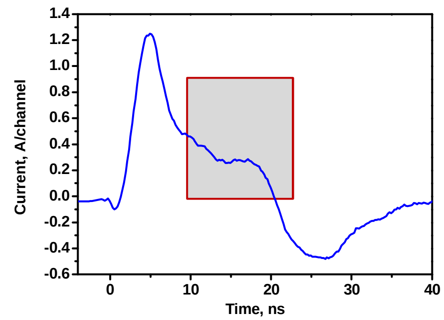
$$E_{ch} = 2U/L_{max}$$



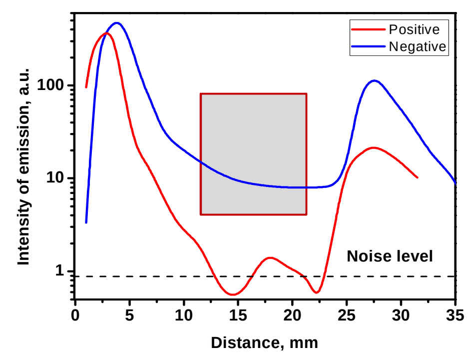


$$\frac{d[N^*]}{dt} = k_{exc} \cdot n_e \cdot [N_2] - [N^*]\left(\frac{1}{\tau_0} + \sum_{i=1} k_q^i [M_q^i]\right) = 0$$

$$n_e = \frac{I}{v_{dr} e S}$$



$$[N_2(C)] = [N_2] \cdot \frac{k_{exc} \cdot n_e}{1/\tau + \sum_{i=1} k_q^i [M_q^i]},$$



Logics of data treatment

LPP

Current and radius measurements $I(t)$, r

E/N on the segment |AB|, $E=U/L_{max}$

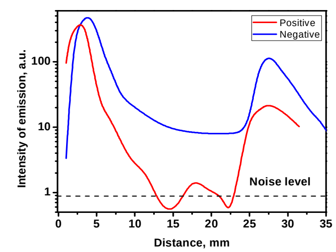
Emission intensity measurements

Electron density n_e on the segment |AB|

Absolute value of $N_2(C)$ on the segment |AB|

Temporal profile of $N_2(C)$ concentration

$f(E/N)=E\mu/k_{exc}=j(t)/k_{exc}n_e$

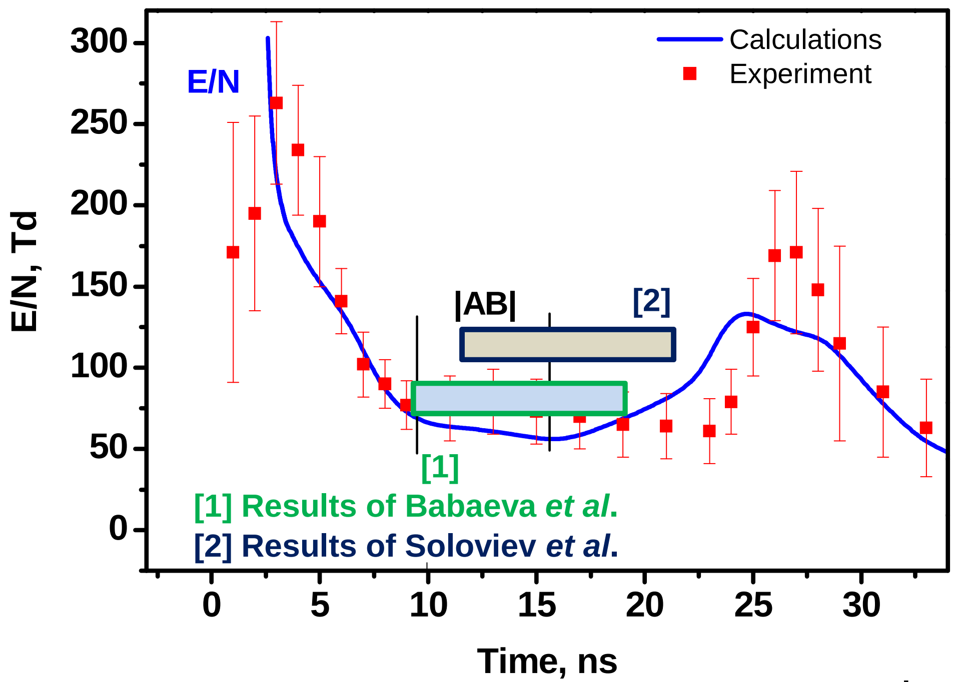


Electron density $n_e(t)$

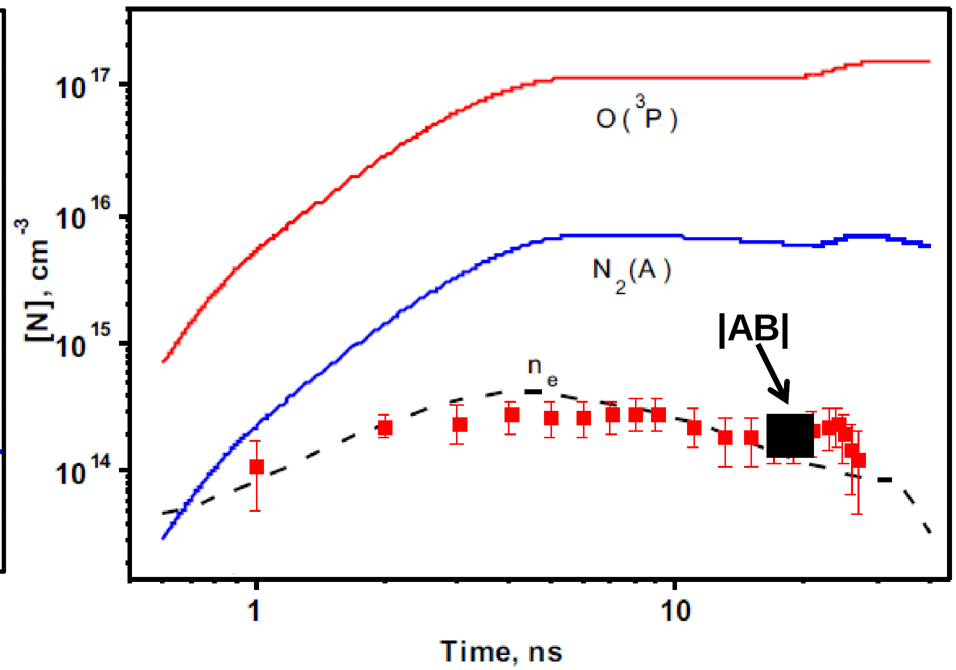
Temporal profile of E/N

Comparison with results of numerical calculations

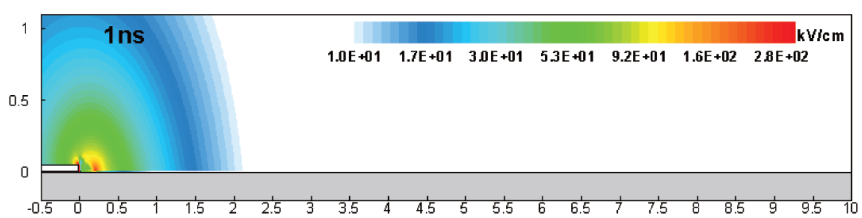
Electric field

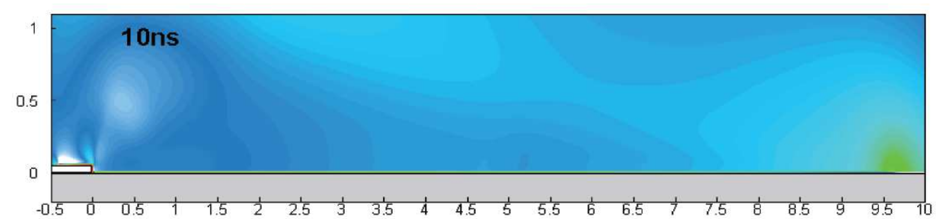


Electron density



Lines: 1D kinetic modeling, N.A.Popov (MSU)





Streamer discharge (U=+10 kV in the cable, T=30 ns)

nSDBD in air at 1 atm:

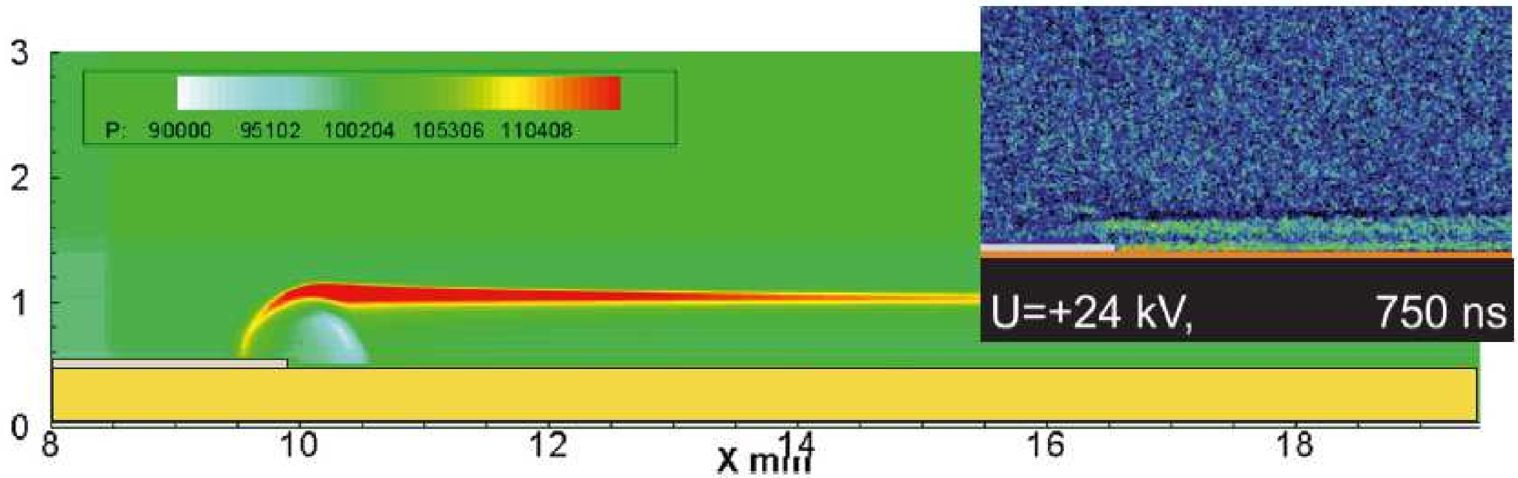
I(t): 0.2 A/streamer (600 A/cm^2);

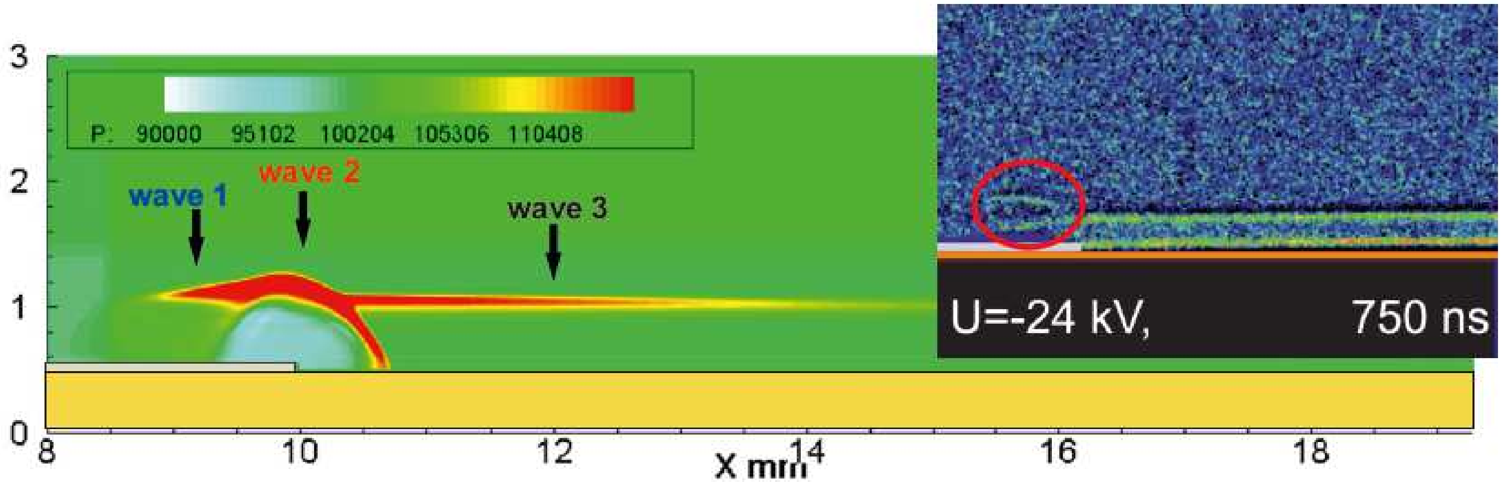
v(t,x): 2-5 mm/ns;

E/N(t,x): 100 Td after the front for U<0, <<100 Td for U>0;

n_e(t,x): 10^{14} cm^{-3}

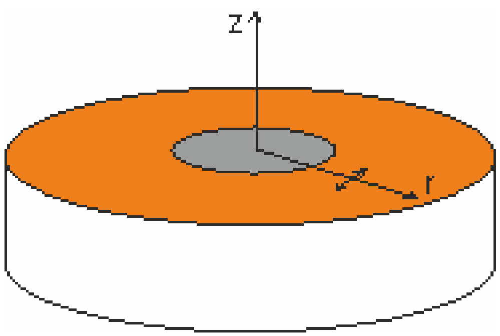
Flow control:
2D modeling and experiments

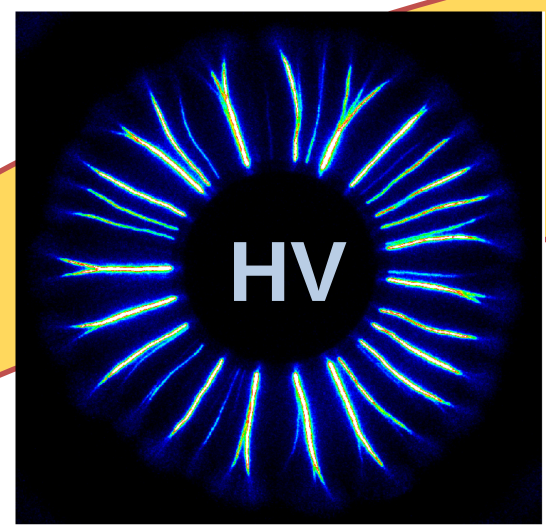


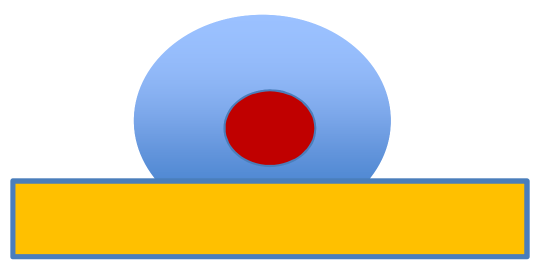


Scale of specific deposited energy

High pressure filamentary SDBD?

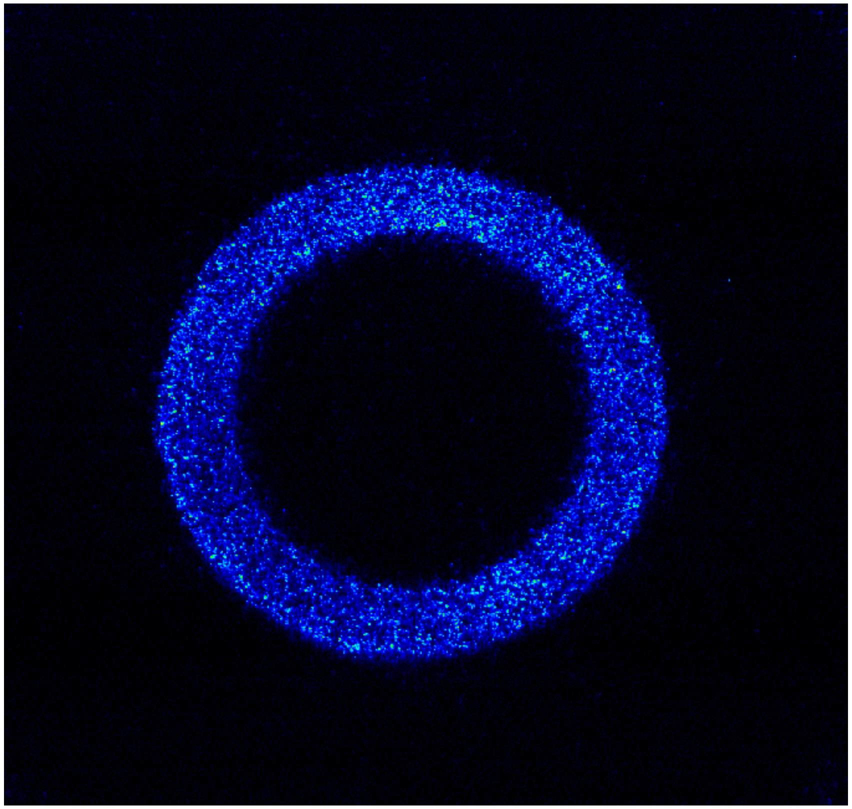




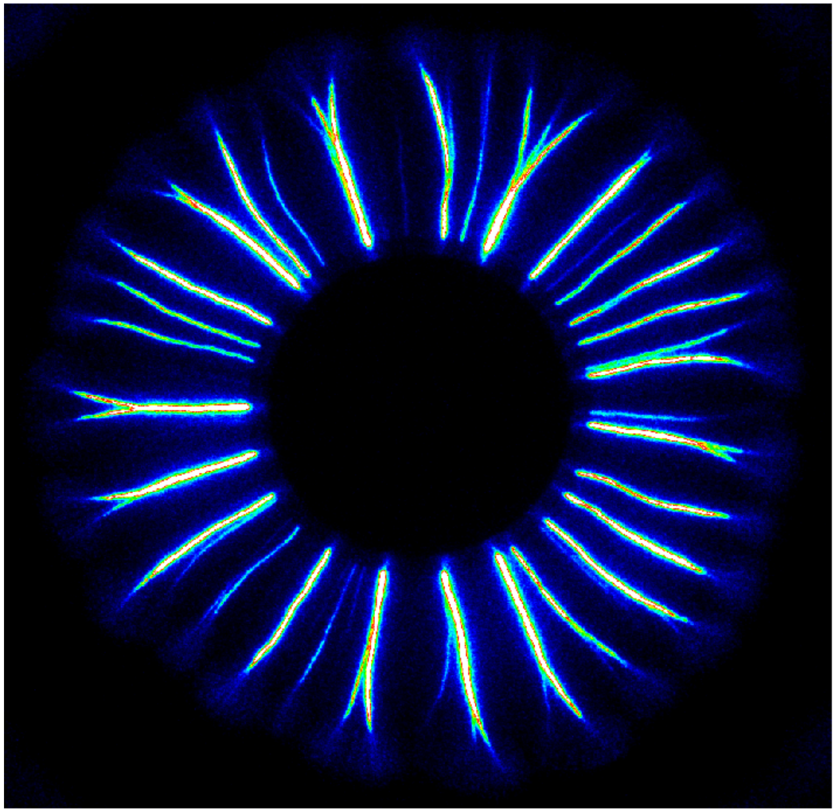


High pressure nSDBD: streamer-to-filament transition

Intensity (filament)=50 x Intensity (streamer)

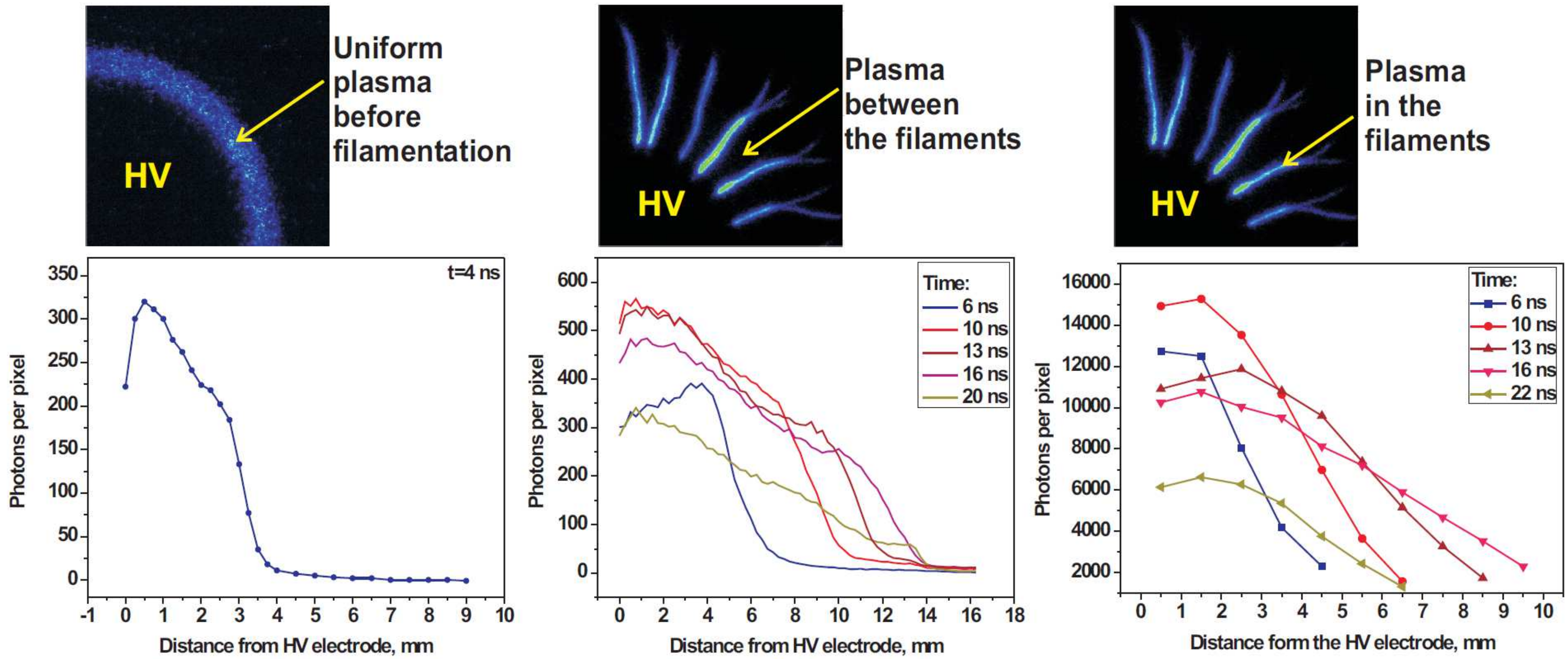


-35 kV, 2 bar, air



-46 kV, 4 bar, air

Analysis of intensities (streamer vs filamentary mode)

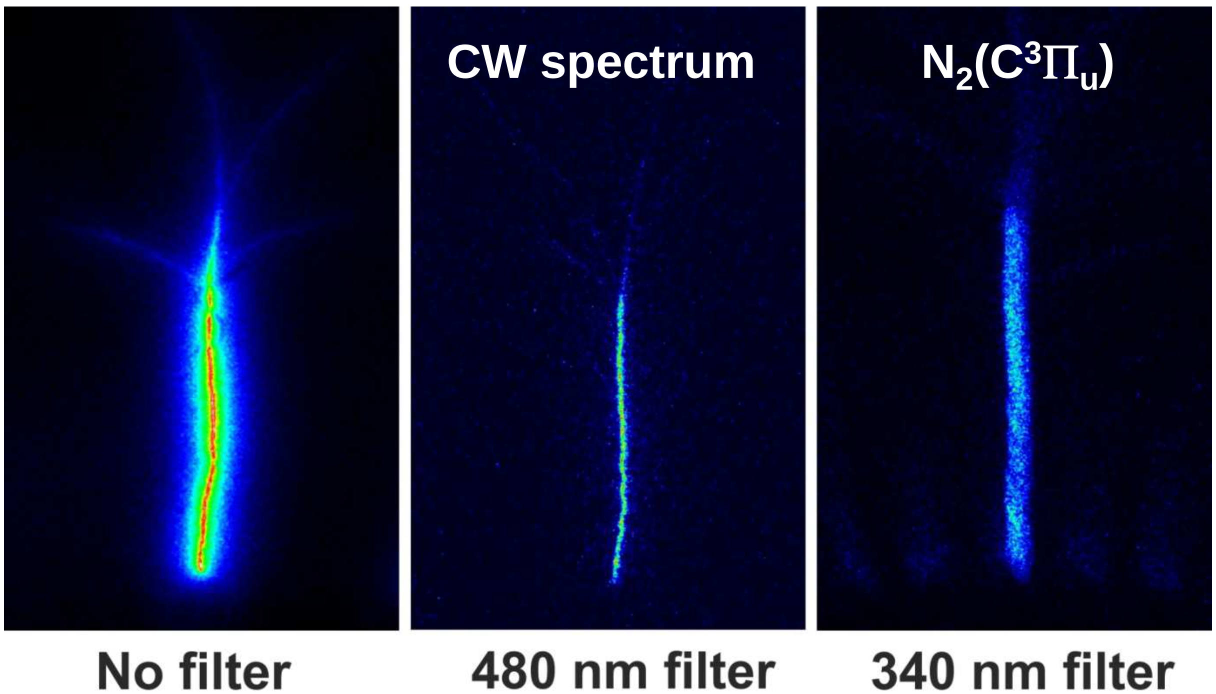


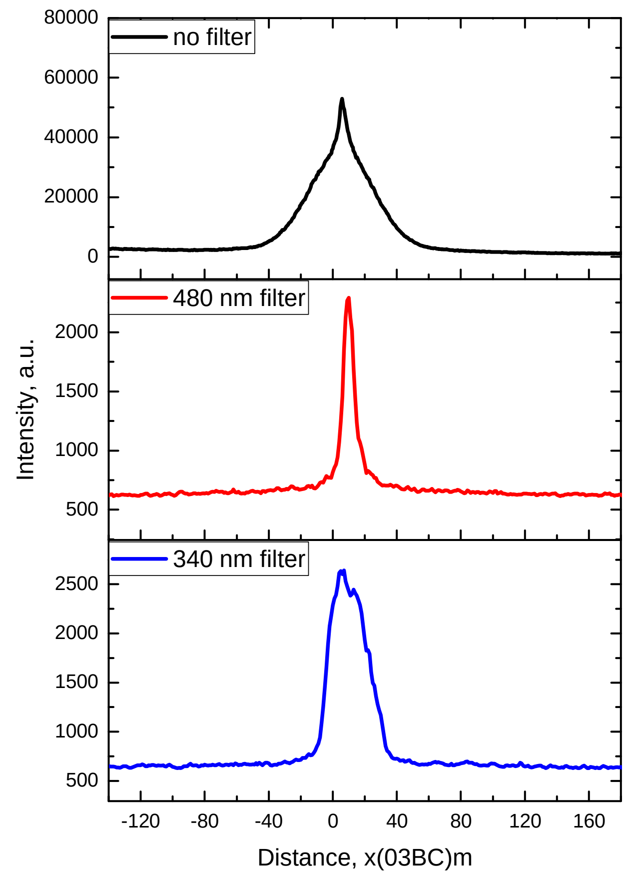
$$I_{11ns}^{fil}/I_{11ns}^{dif}/I_{4ns}^{dif} \sim 50/1.5/1$$

Radial distribution of the frontal discharge emission; P=3 bar, U=-46 kV

Optical emission in the filaments

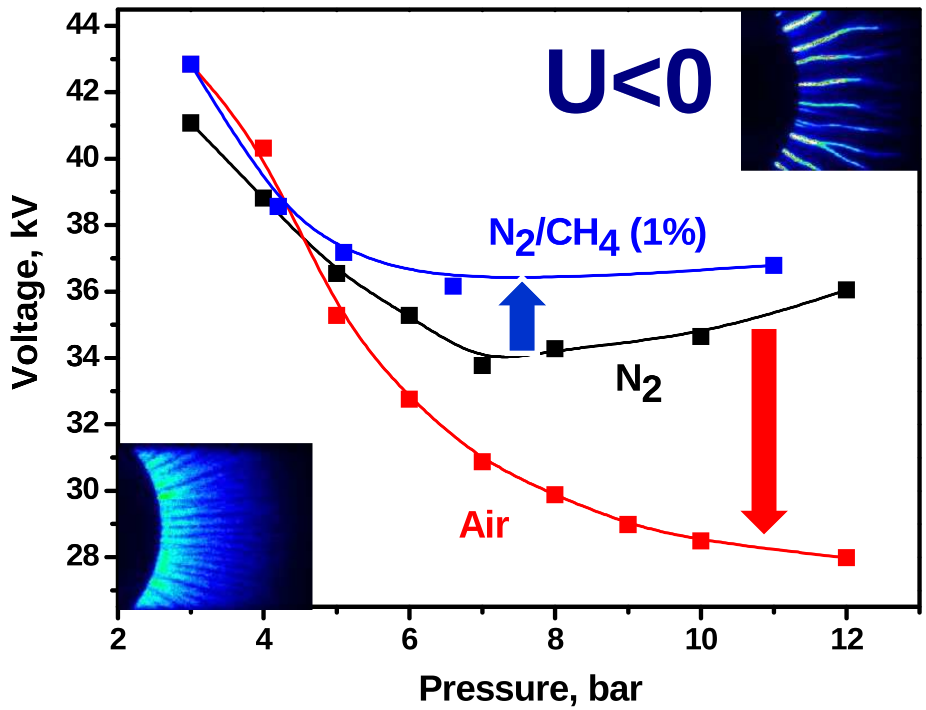
ICCD images and radial intensity profiles of single filament: without filter and with (480±5) nm and (340±5) nm narrowband filters

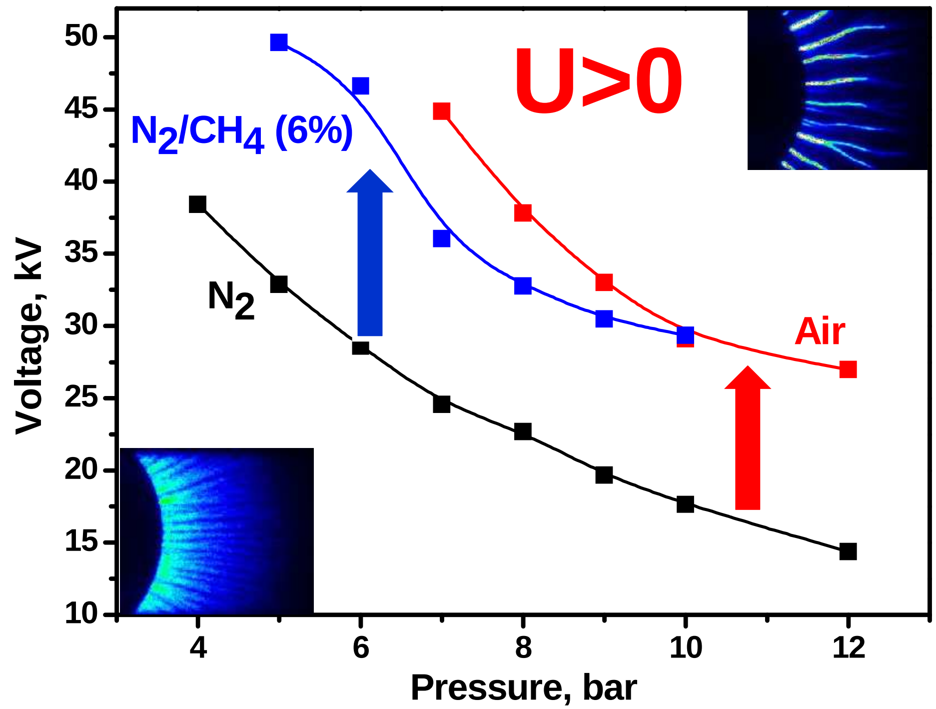




ICCD camera gate is 10 ns

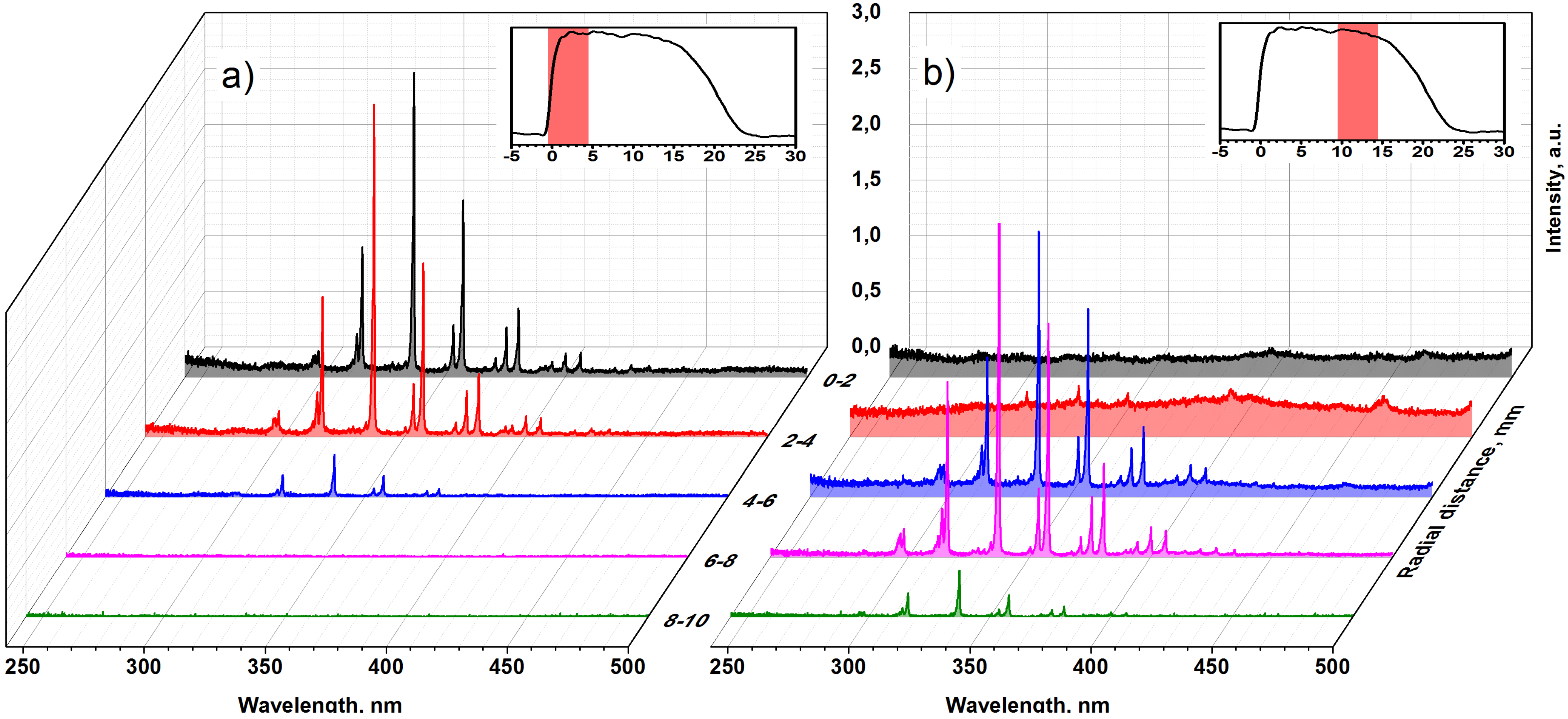
Filamentation curves for nitrogen, air and N_2/CH_4 mixtures



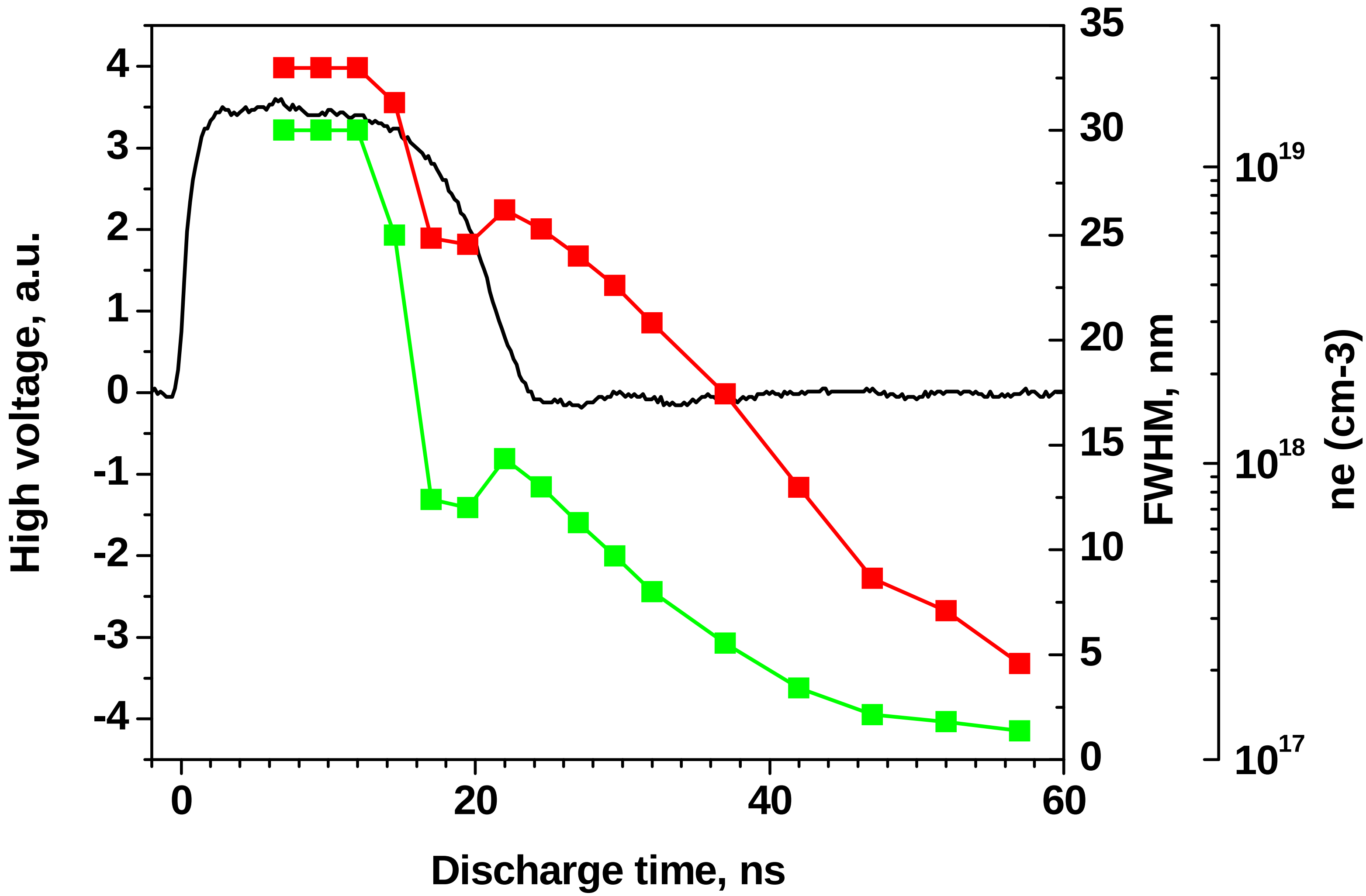


Continuous spectra in filamentary discharge

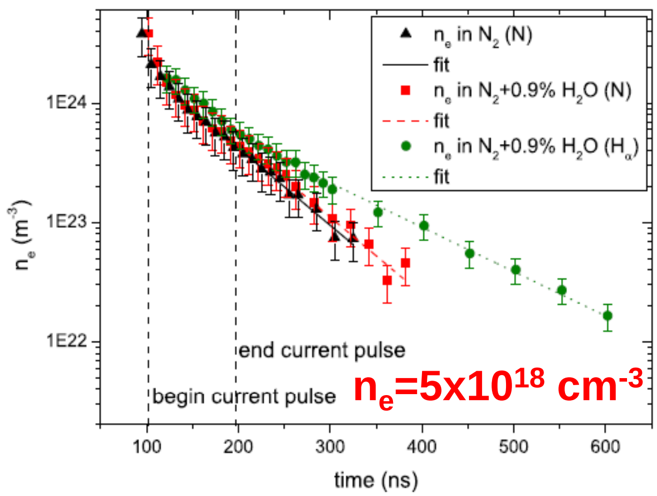
LPP



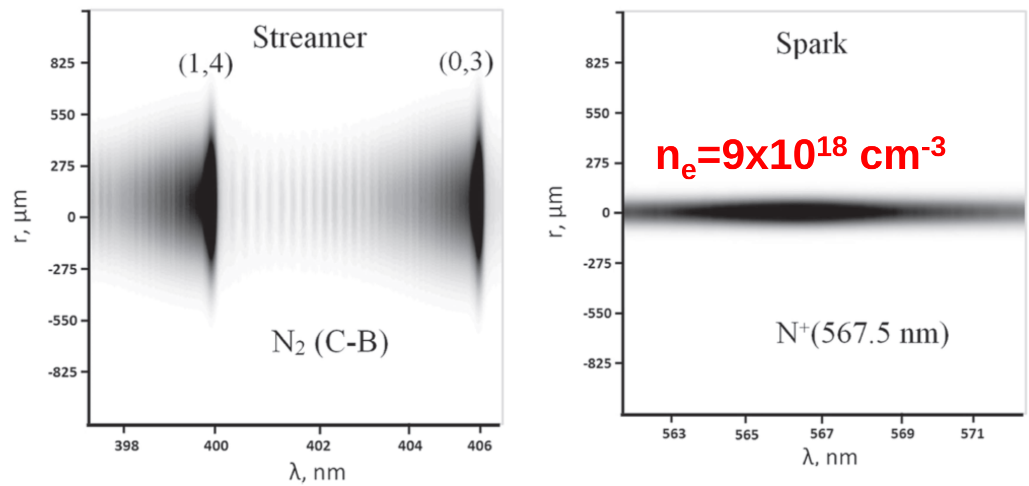
Electron density in the filaments: H-atom emission at 656 nm



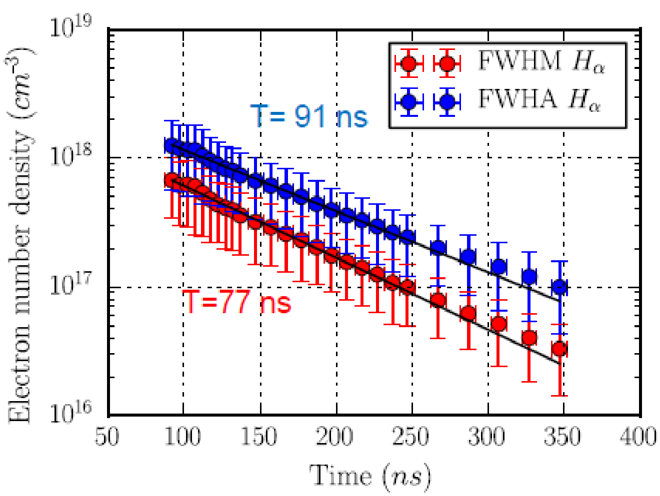
Group of support: for high electron densities!



Van der Horst R., Verreycken T, Bruggeman P. et al J. Phys.D. (2012) 345201

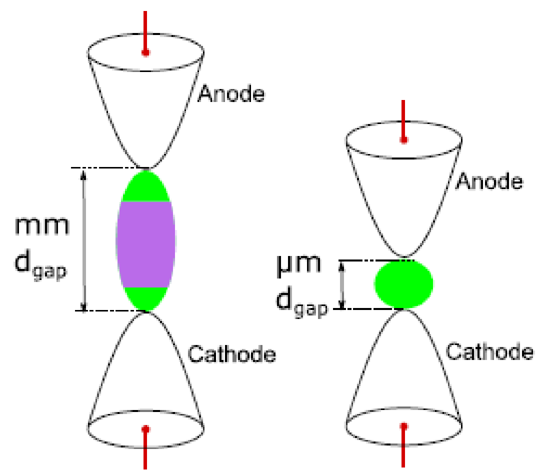


Lo A., Cessou A, Lacour C. et al PSST (2017) 045012

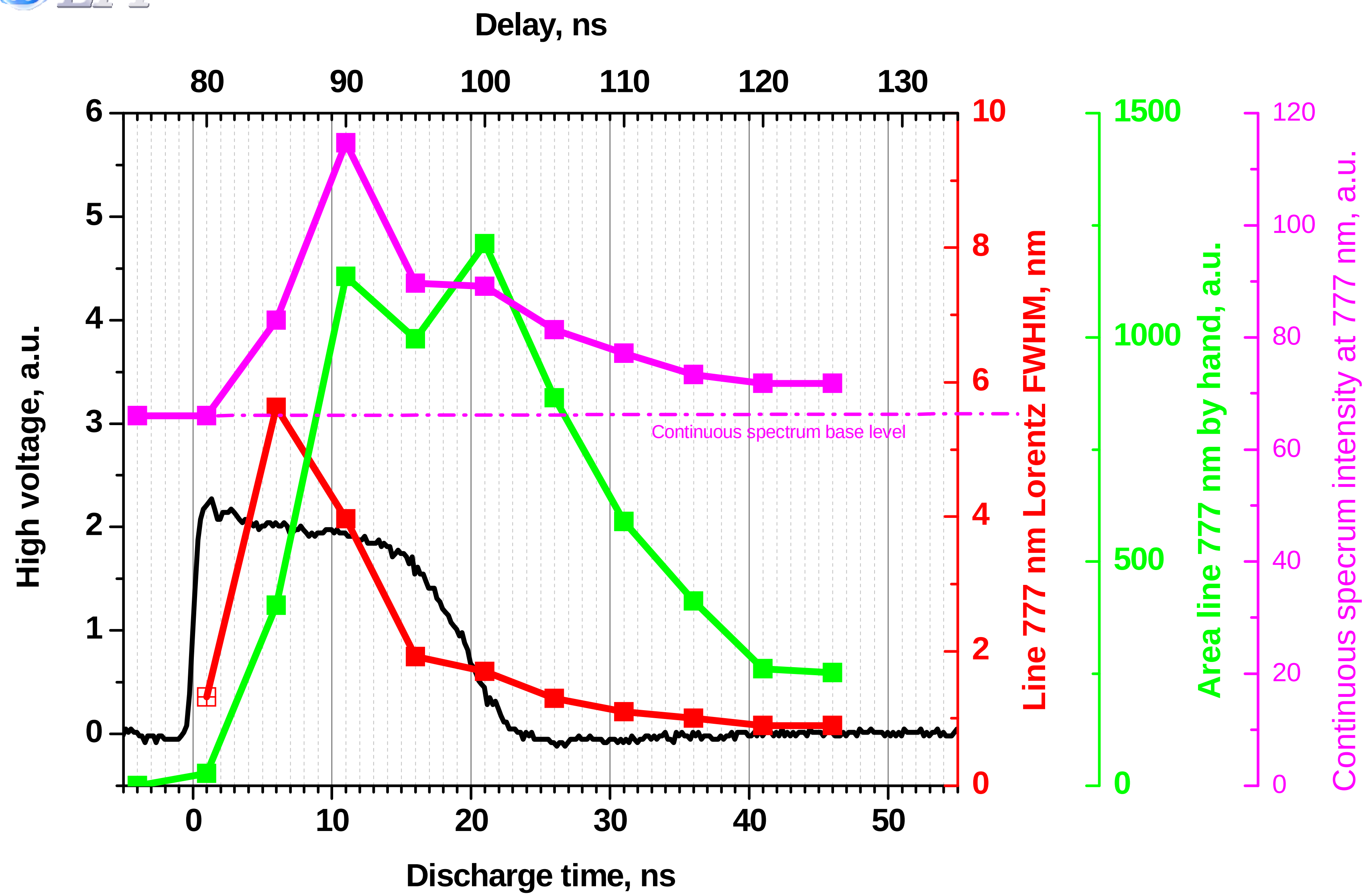


$n_e=1.5x10^{18}$ cm^{-3}

D. Pai, GEC 2016



Correlation in time: U-pulse, FWHM of O-line, intensity of CW emission, O* –atom density

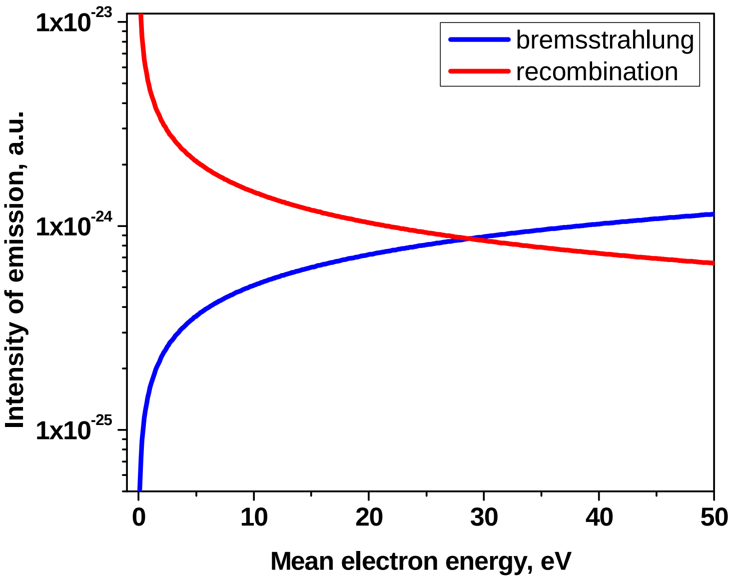


Possible reasons of CW spectrum-1

Bremsstrahlung and recombination radiation

$$Q_{br} \propto N_e \cdot N_{ion} \cdot \sqrt{T_e}$$

$$Q_{rec} \propto \frac{N_e \cdot N_{ion}}{\sqrt{T_e}}$$



Intensity of bremsstrahlung and recombination radiation is

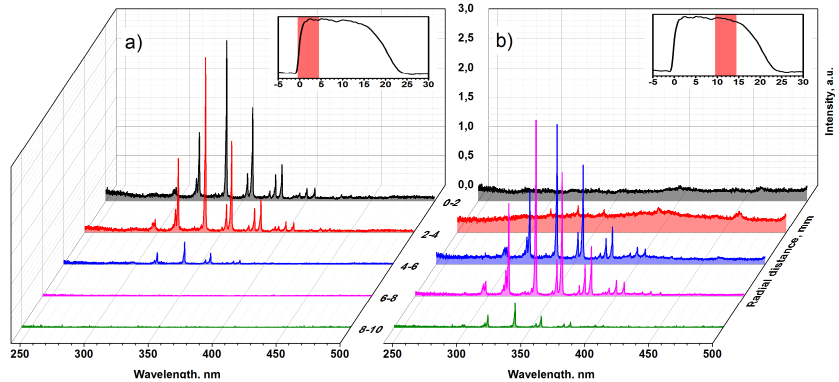
a strong function of density of charged species

and weakly depends on the electron temperature

Possible reasons of CW spectrum-2

SPS (337.1 nm) is a reference radiation

$$\begin{cases} N_2(C) = k_C(E/N)\cdot n_e \cdot N_2/\, v_q \\ Q_C = N_2(C)\cdot F_{FK}\cdot A_{00} \\ Q_C = 3\cdot 10^{22} \text{ quantum/cm}^3\text{/s.} \end{cases}$$

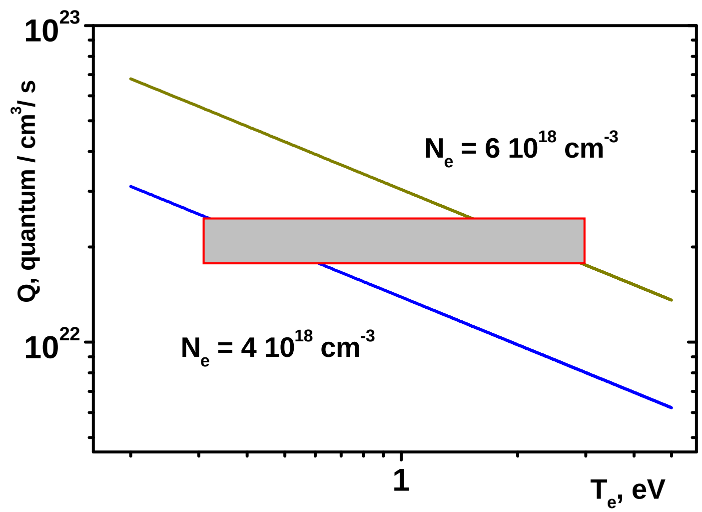


$Q_C(t=0\text{ ns})/Q_{CW}(t=5\text{ ns})\sim(1.5\text{-}2)$

Electron density from recombination emission:

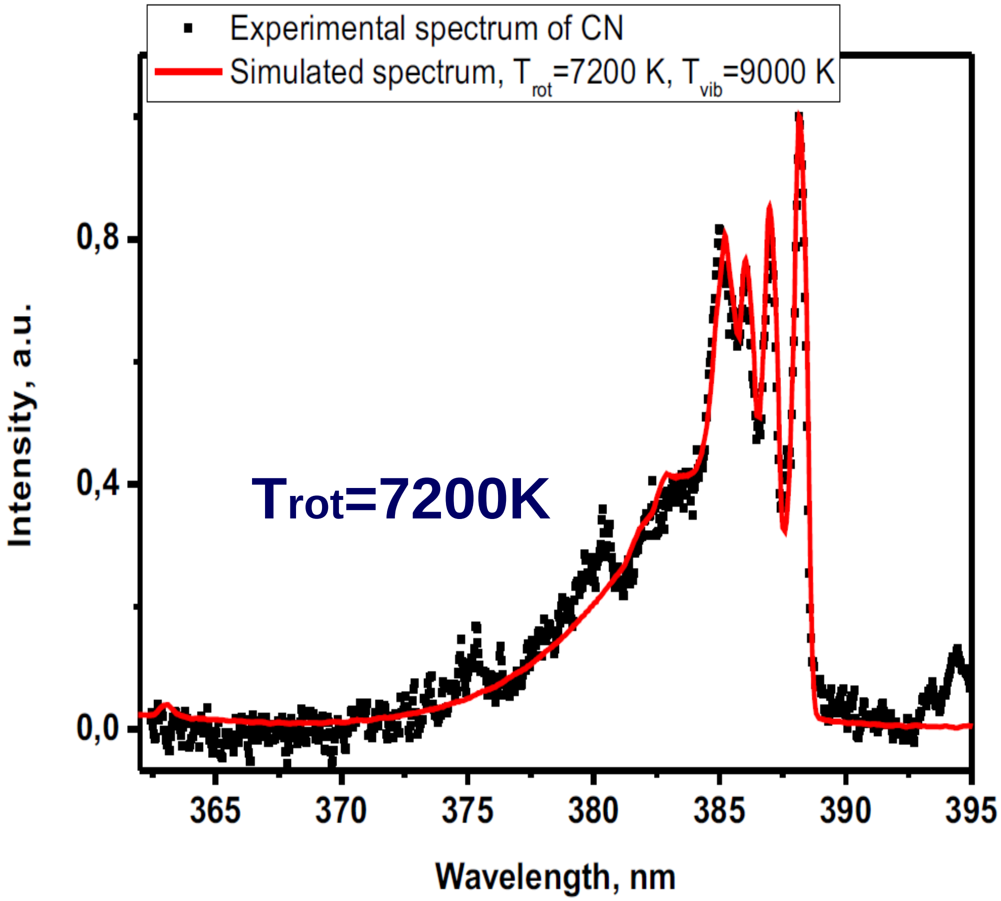
$$Q_{CW}(\omega) = C_0 \cdot \frac{n_e \cdot n_{ion}}{\sqrt{T_e}\cdot \eta\omega} d\omega = 2\cdot 10^{22} \text{ quantum/cm}^3\text{/s}$$

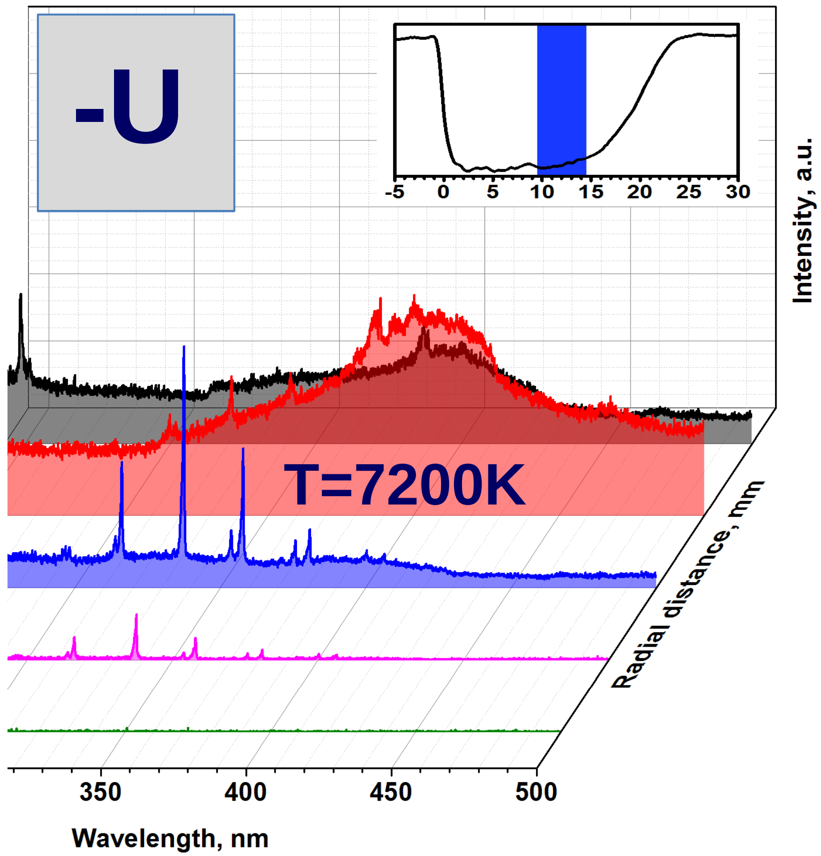
At λ = 337 nm and $\Delta\lambda$ = 3 nm, $d\omega = 5\cdot 10^{13}$ s^{-1}



In the discharge T_e~2-3 eV → n_e~ $6\cdot 10^{18}$ cm^{-3}
In the afterglow T_e~0.5 eV → n_e~ $4\cdot 10^{18}$ cm^{-3}

Continuous spectra in filamentary discharge: Planck emission so gas heating?





The most typical features of the filament

-the electron density is extremely high, 10^{18}-10^{19} cm^{-3} at 6 bar, but the decay is slow, tens of ns;

- no molecular emission is observed from the filament, only cw and atomic lines (no material of electrodes, no energetic ion lines);

-

- observed cw spectrum is due to recombination and Planck emission;

- there is strong (kK) and fast (units of ns) gas heating within the filament

Each feature needs additional analysis: molecular emission

-no molecular emission is observed from the filament, only cw and atomic lines

Quenching by electrons?

$n_e=10^{19}$ cm^{-3},
$k_q=10^{-7}$ cm^3/s

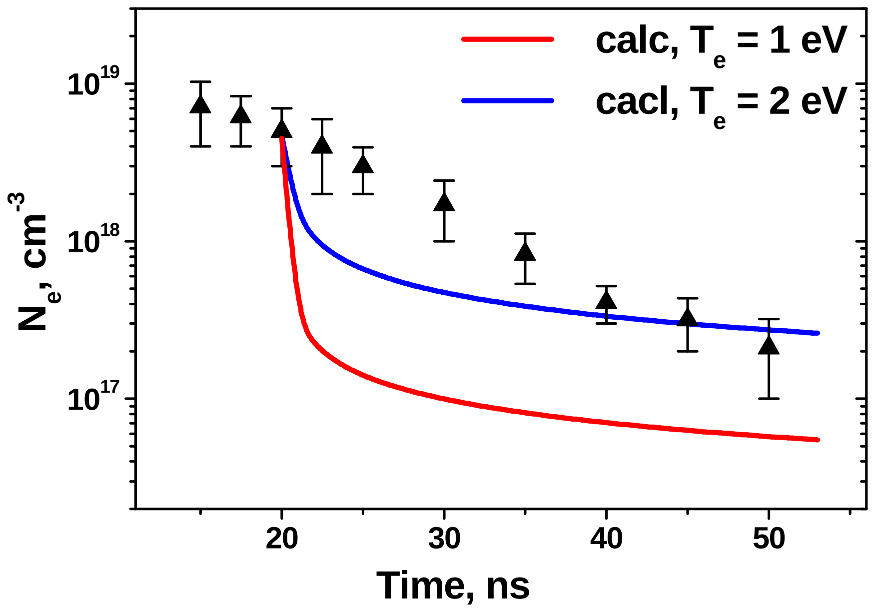
No molecules, 100% dissociation

$N_2^+ + e = N + N$,
$kn_e^2 t > [M]$ during 1 ns

Each feature needs additional analysis: decay time for electrons

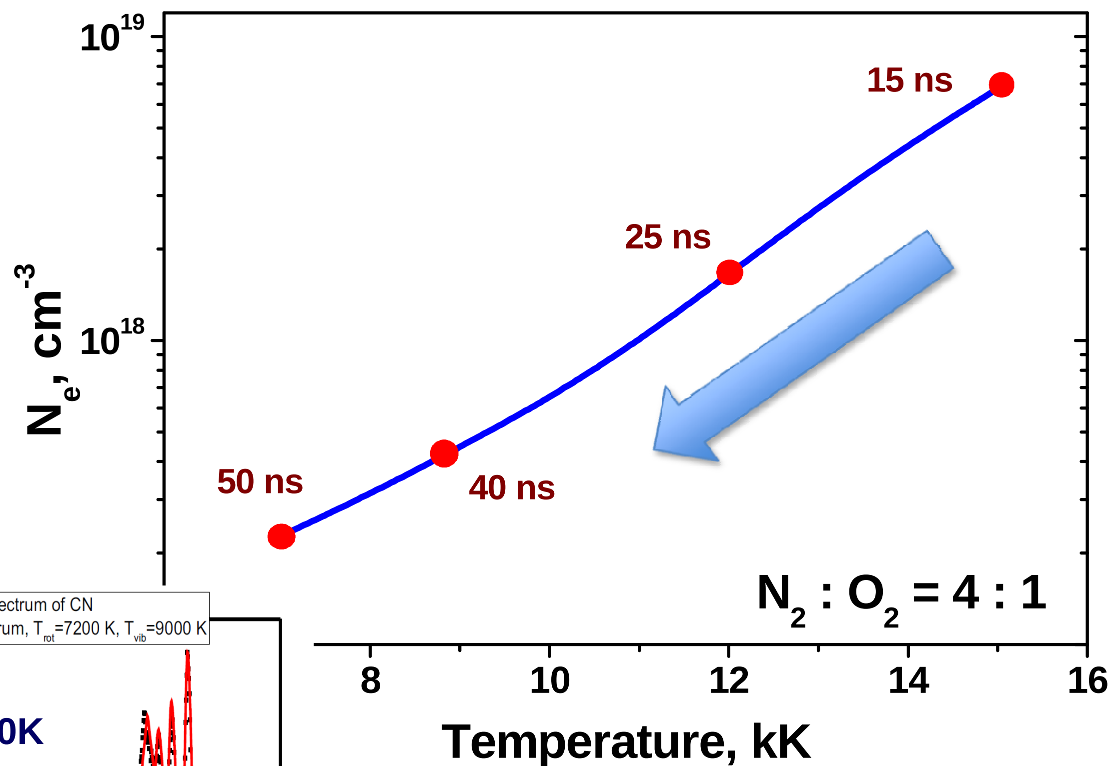
-the electron density is extremely high, 10^{18}-10^{19} cm^{-3} at 6 bar, and the decay is low, 10s of ns

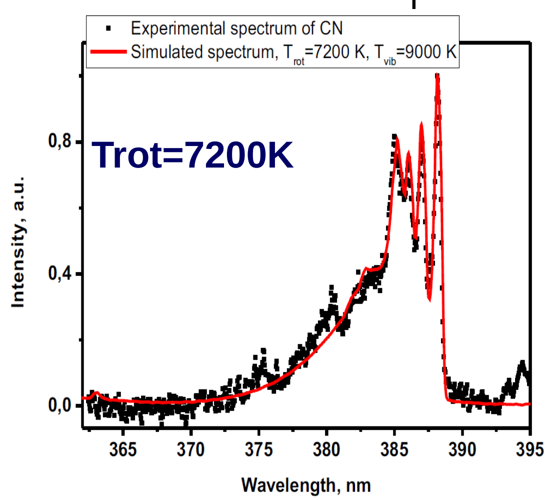
Recombination decay, N^{+}+e+e is too fast (parts of nanoseconds)



Thermal ionization?

Each feature needs additional analysis: decay time = gas cooling for electrons?

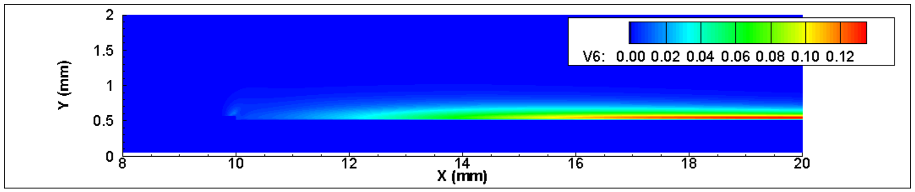




Thermal ionization?

And finally, how this transition starts?

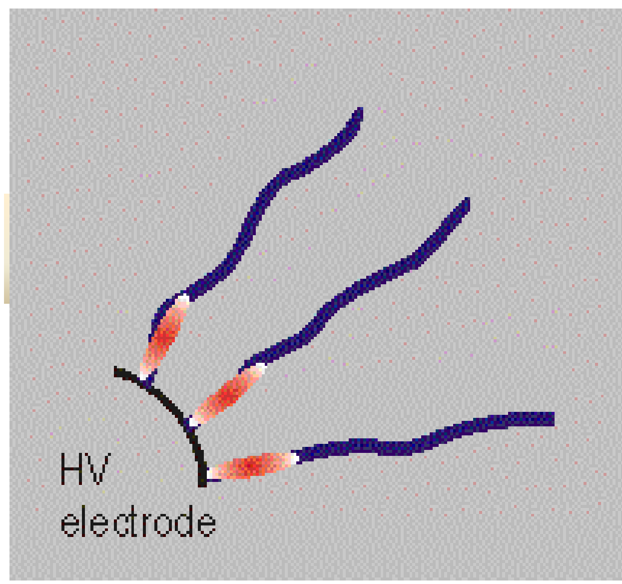
Increase of T by kinetic FGH



$T \Uparrow$

Ionization-heating instability?

Decrease of gas density within a few μm distance



$N \Downarrow$

$$\tau_p = 1 - 3\, ns$$

Increase of E/N

$$\frac{E}{N} \Uparrow$$

Increase of ionization

Conclusions

Specific deposited energy in nanosecond microplasmas can be as high as electronvolts per molecule. At high reduced electric fields and moderate gas pressures, "non-diluted chemistry" in the discharge and early afterglow is important

$N_2(C^3\Pi_u)$ emission in P=1 atm surface streamer discharge, electrical current and morphology provide important information about the plasma parameters

High pressure filamentary nSDBD is characterized by abrupt cw emission and by high electron density. Appearance and physics of streamer-to-filamentary transition are studied

Acknowledgments

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Thank you for your attention