

# Plasma assisted high pressure combustion; surface HP nanosecond DBD



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# Our scientific team for PAI/PAC problem:



NeQ systems Laboratory, Moscow, 1998-2006

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Moscow State University, Skobeltsyn Institute of Nuclear Physics:

Nikolay Popov

PC2A, Lille:

Mohamed Boumehti,  
Guillaume Vanhove,  
Pascale Desgroux



# Plan of the presentation



Introduction: physics and chemistry of plasma-assisted combustion

Shock tube experiments: 0D, low pressures, high temperatures

Rapid compression machine (RCM) experiments: 2D, high pressures, low temperatures

High pressure high temperature (HPHT) discharge cell experiments: the discharge and the following combustion

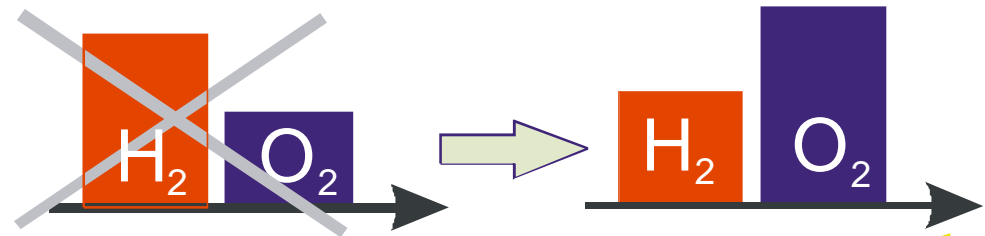
High pressure discharge: streamer-to-filament transition

# I. Physics and chemistry of plasma assisted combustion

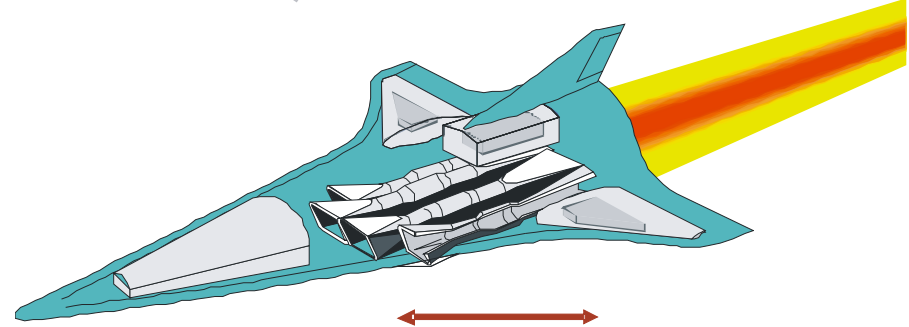
# Plasma assisted ignition/combustion: nonequilibrium plasma applications



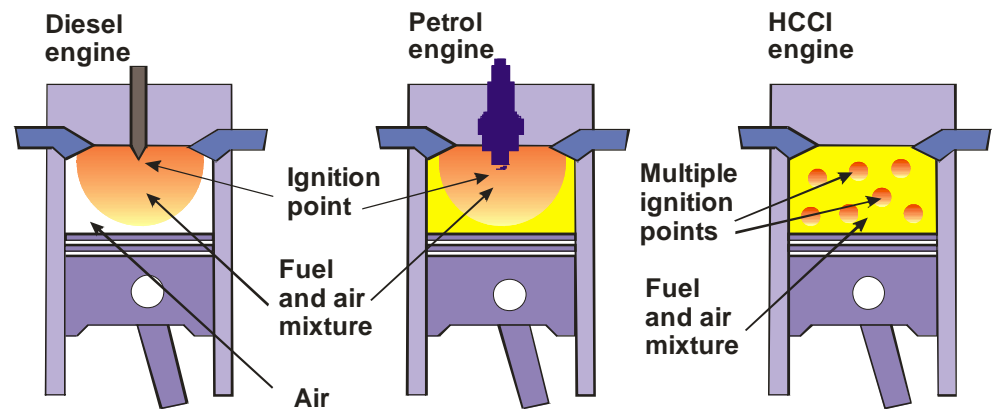
- Lean mixtures



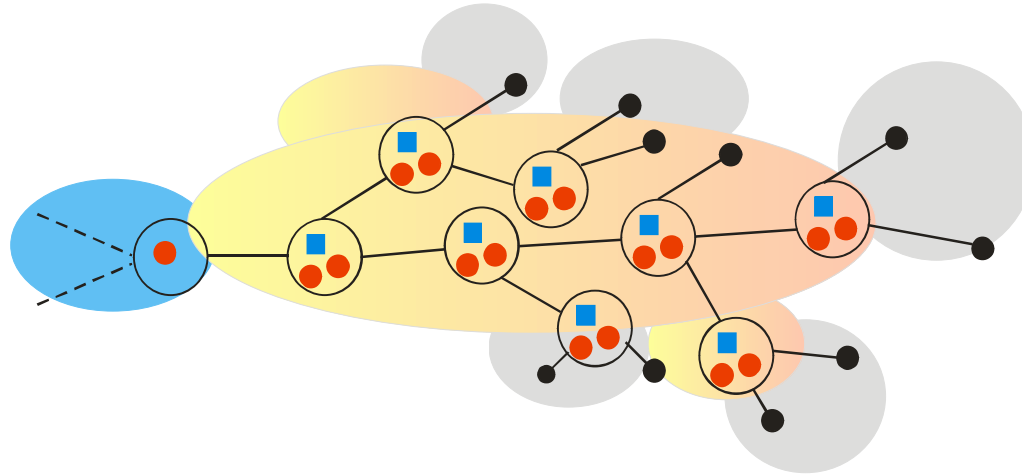
- Fast flows (1998)



- High pressures

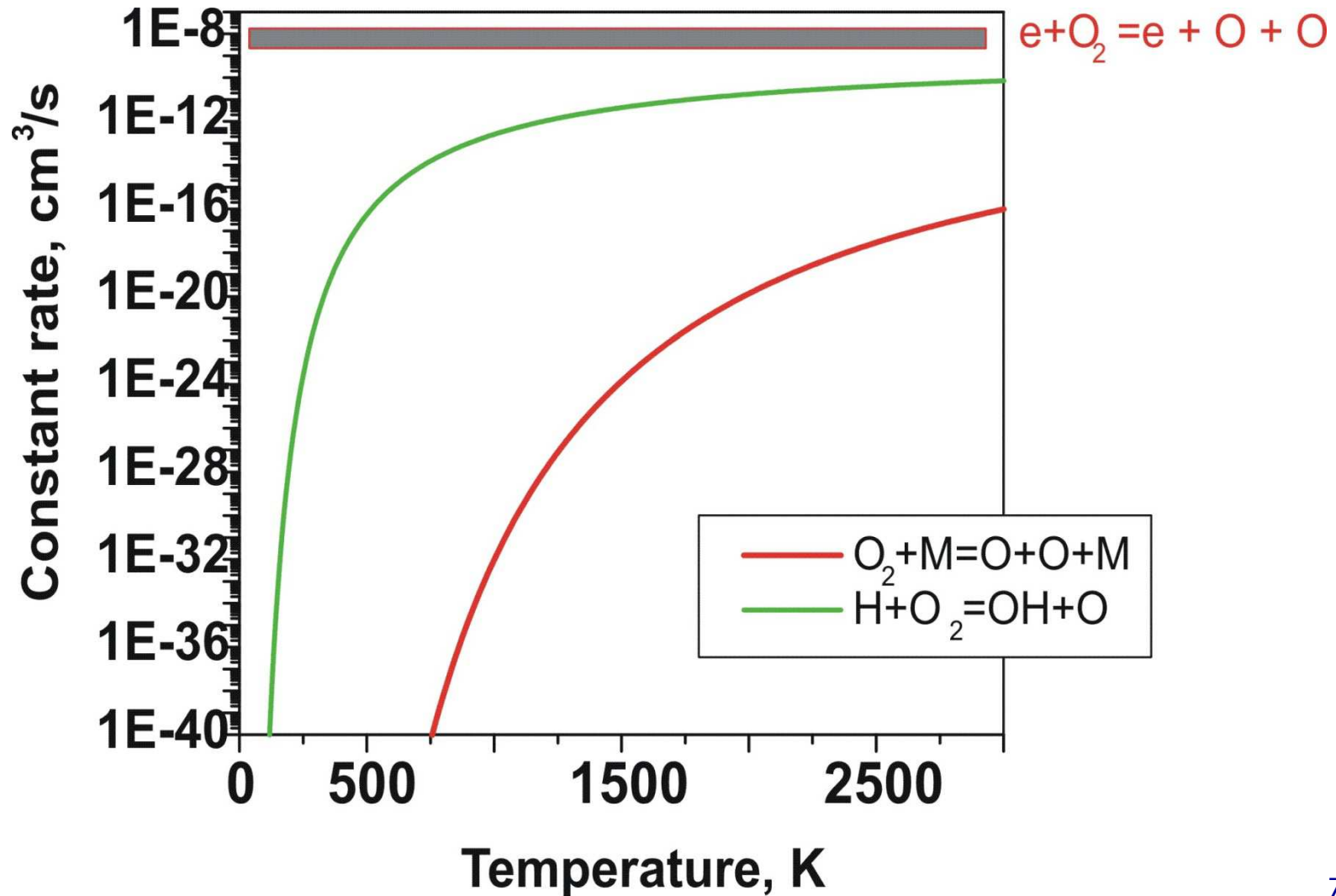


# Combustion: chain reactions

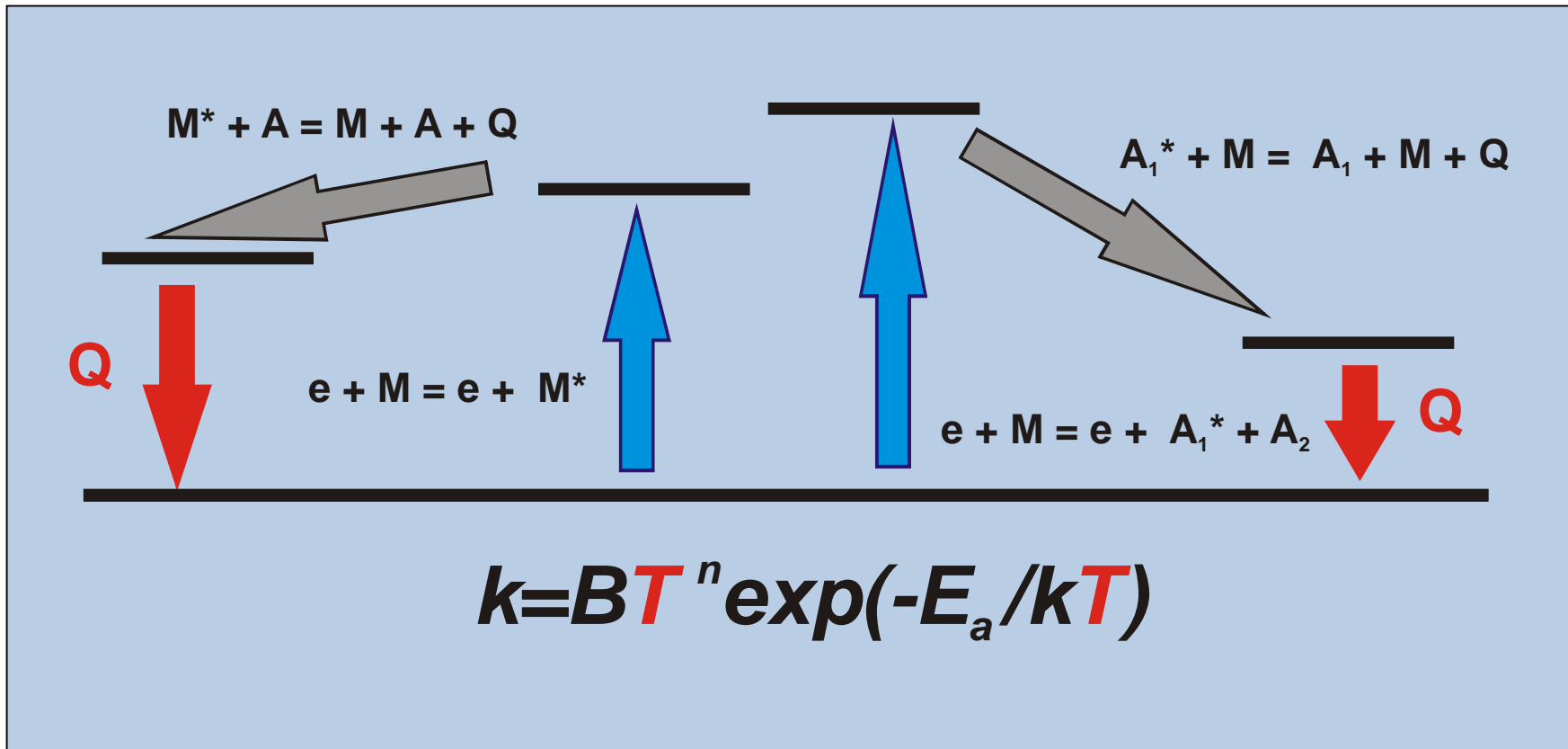


Initiation	$\text{H}_2 + \text{O}_2 = 2 \dot{\text{O}}\text{H} + 78 \text{ kJ}$
Branching	$\dot{\text{H}} + \text{O}_2 = \dot{\text{O}}\text{H} + \dot{\text{O}} + 70 \text{ kJ}$ $\dot{\text{O}} + \text{H}_2 = \dot{\text{O}}\text{H} + \dot{\text{H}} + 8 \text{ kJ}$
Development: 2x	$\dot{\text{O}}\text{H} + \text{H}_2 = \text{H}_2\text{O} + \dot{\text{H}} - 62 \text{ kJ}$
Break	$\dot{\text{H}} + \text{wall}$ $\dot{\text{H}} + \text{O}_2 + \text{M} = \text{H}\dot{\text{O}}_2 + \text{M} - 203 \text{ kJ}$

# Comparison of the reaction rates



# Fast gas heating: time less than VT-relaxation

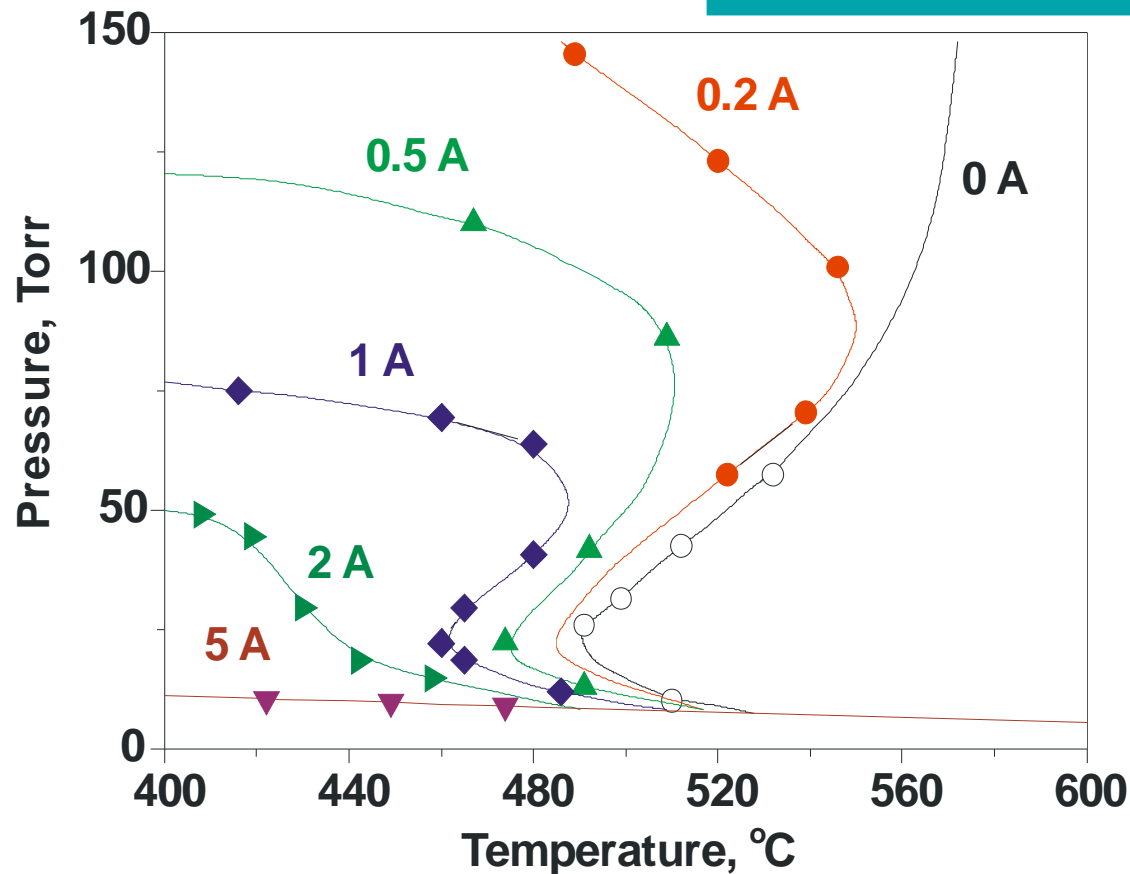




# First available experiments on plasma-assisted combustion

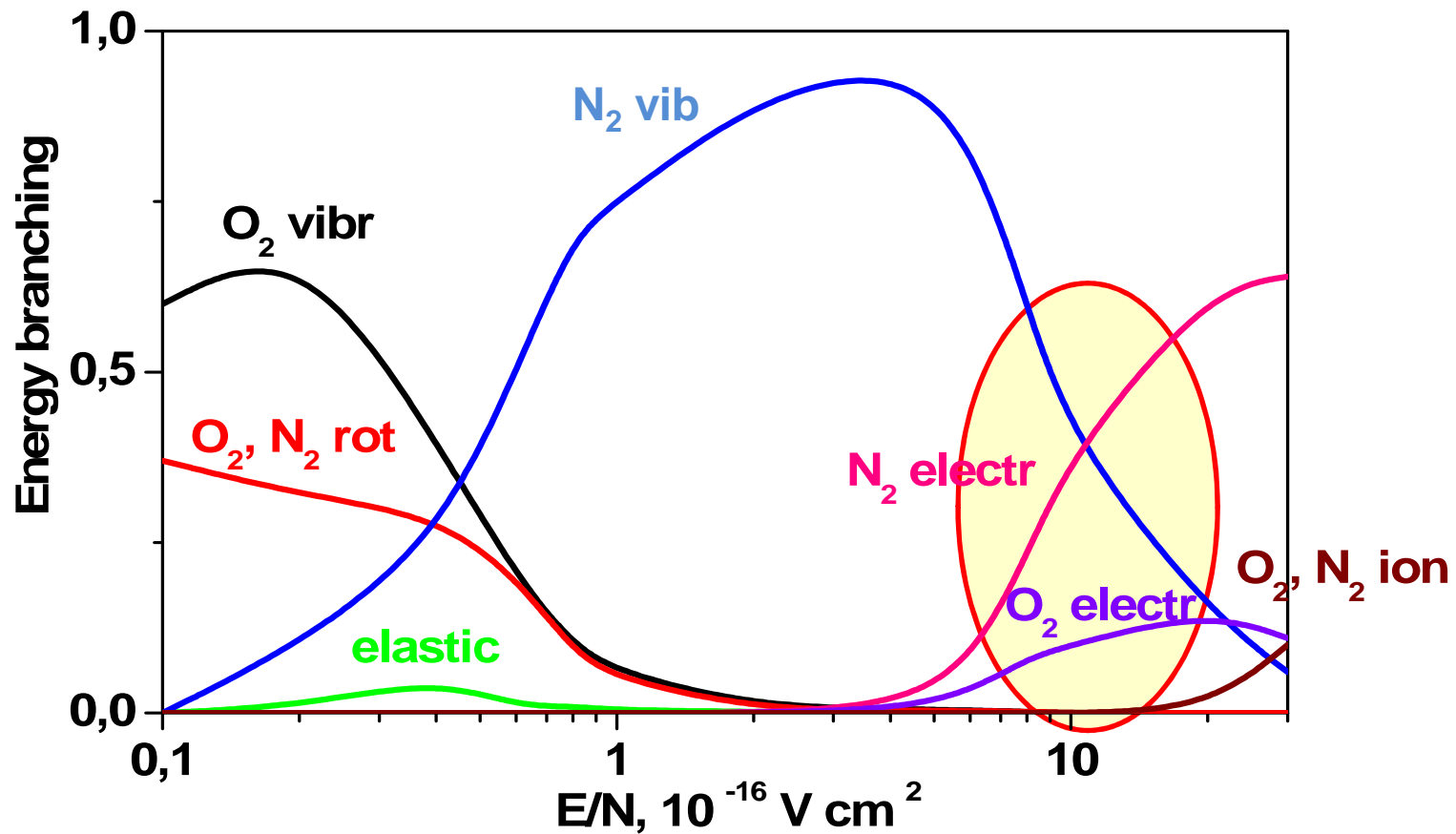


Semenov N N (1958) Some problems of chemical kinetics and reactivity Pergamon Press



G.Gorchakov, F.Lavrov, Influence of electric discharge on the region of spontaneous ignition in the mixture  $2\text{H}_2\text{-O}_2$ . Acta Physicochim. URSS (1934), 1, 139-144

# Energy branching vs reduced electric field $E/N$

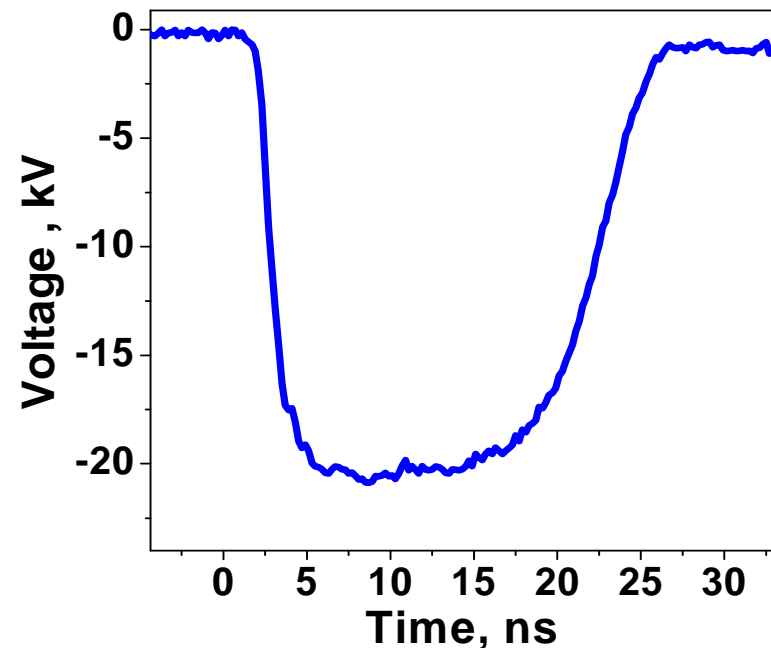
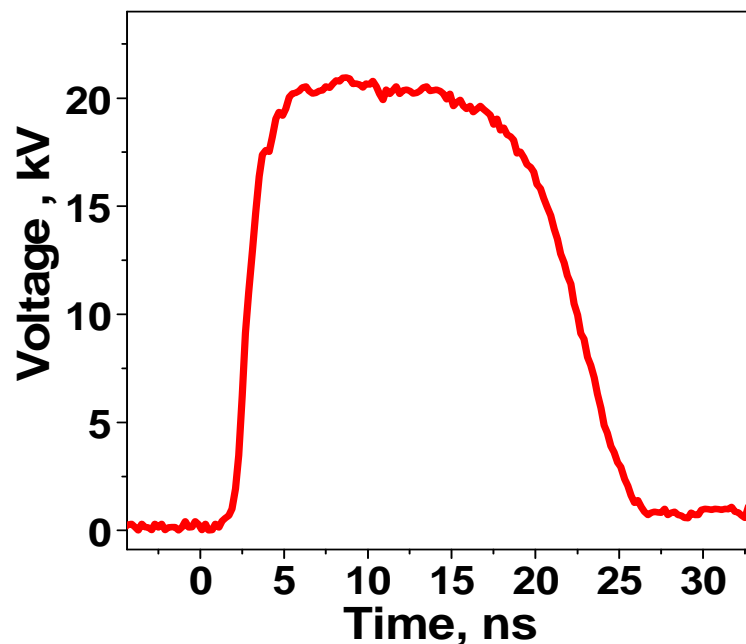


N.L. Aleksandrov, E.E. Son, 1980, in: *Plasma Chemistry*, B.M.Smirnov, ed., Atomizdat Publ., Moscow, V.7, pp. 35-75

# Applied high voltage pulses

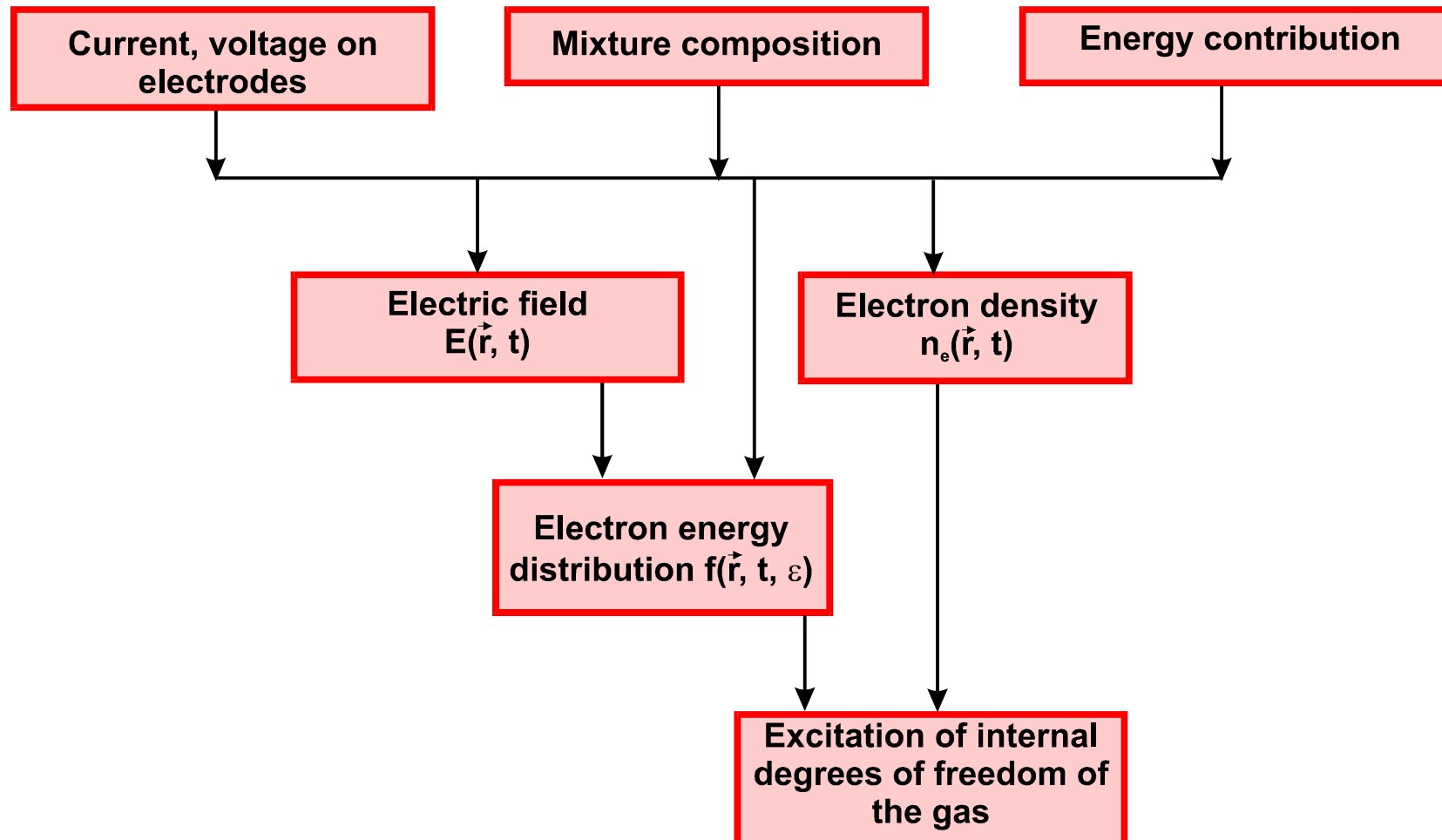


## Positive and negative polarity pulses

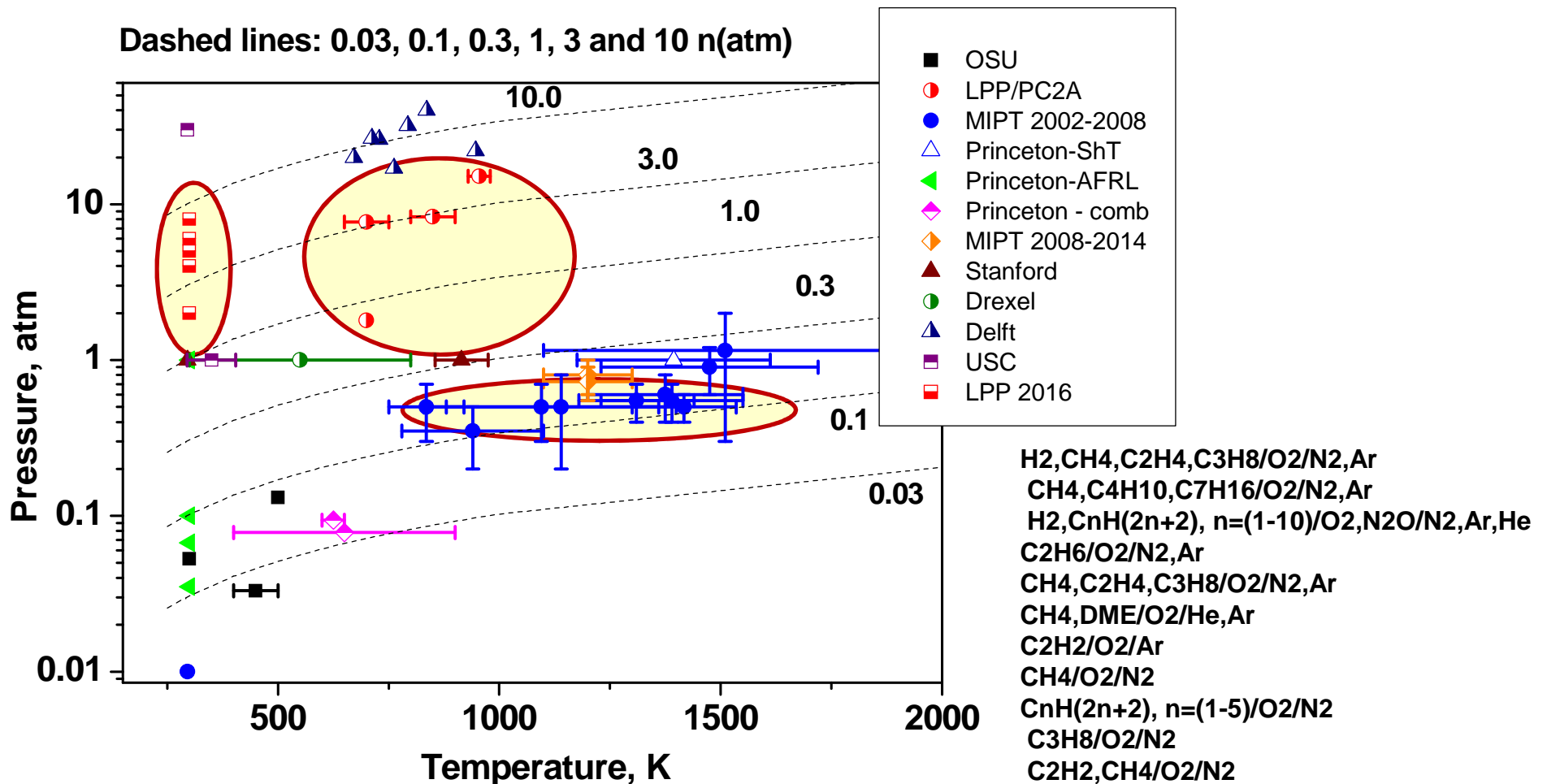


Single pulse regime, 20 ns pulse duration, 2 ns front rise time

# Kinetic approach to description of a gas discharge



# Field of parameters where nanosecond PAI/PAC experiments are available



S M Starikovskaia, J. Phys. D: Appl. Phys., 47 (2014) 353001 (34pp)

**All experiments were performed in a  
SINGLE-SHOT regime**

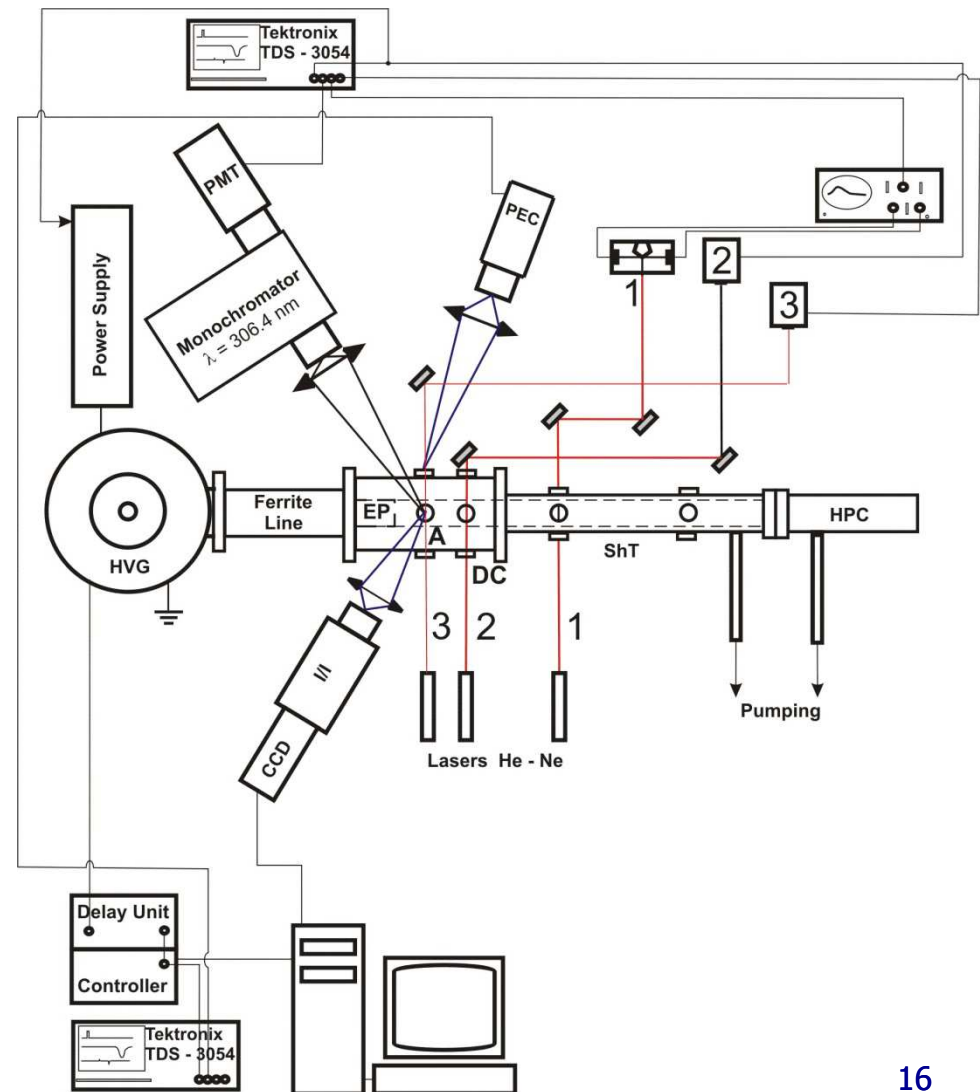


## II. Shock tube experiments: [relatively] low $P$ and high $T$

# Shock tube setup for plasma ignition

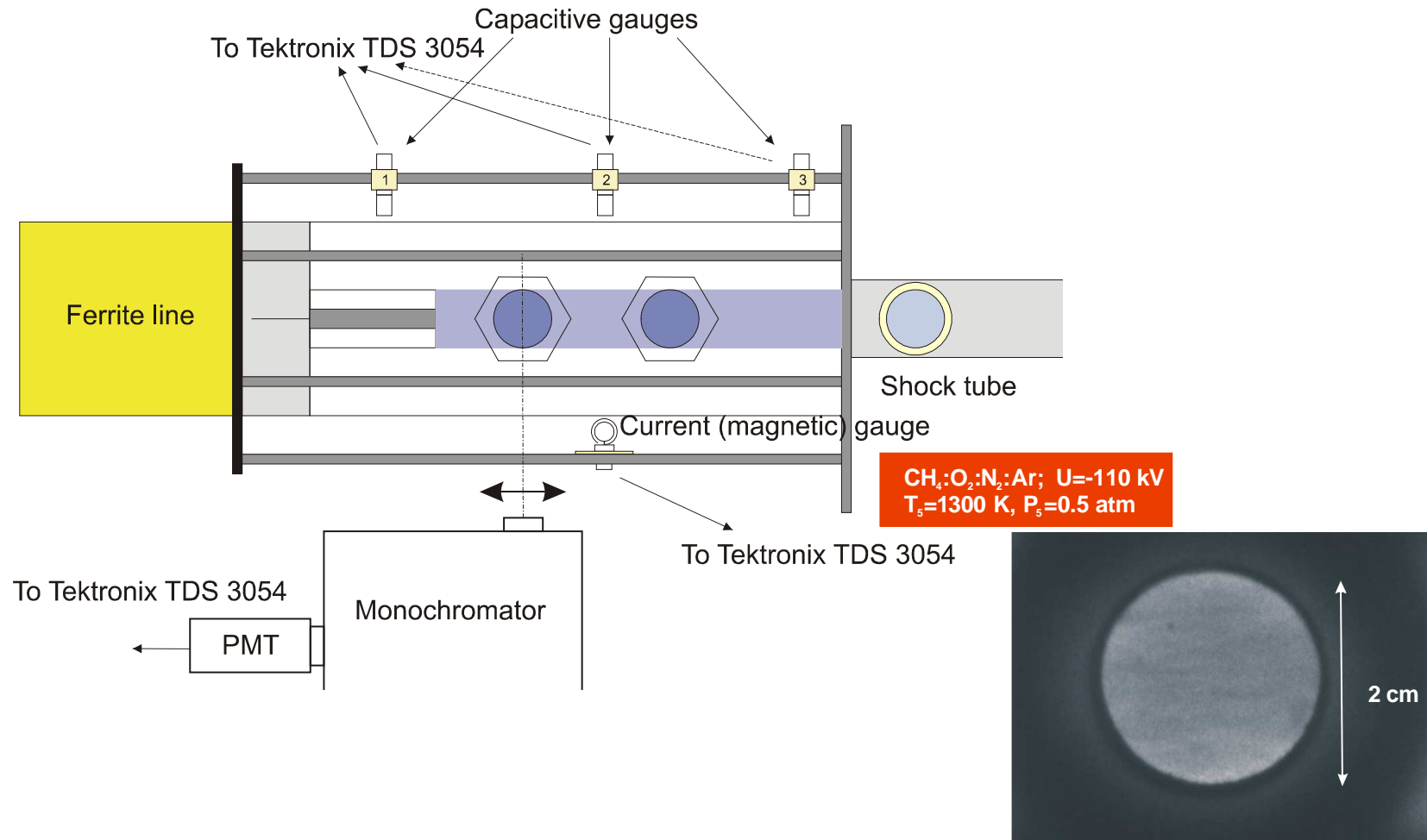


- Mixture composition  
 $\text{CH}_4/\text{C}_2\text{H}_6/\text{C}_3\text{H}_8/\text{C}_4\text{H}_{10}/\text{C}_5\text{H}_{12}$   
-  $\text{O}_2$  - Ar (90%)
- Temperature  
950-2000 K
- Pressure  
0.2-1.0 atm
- $T_5, P_5$
- $T_{\text{ign}}$
- E/N, I, W



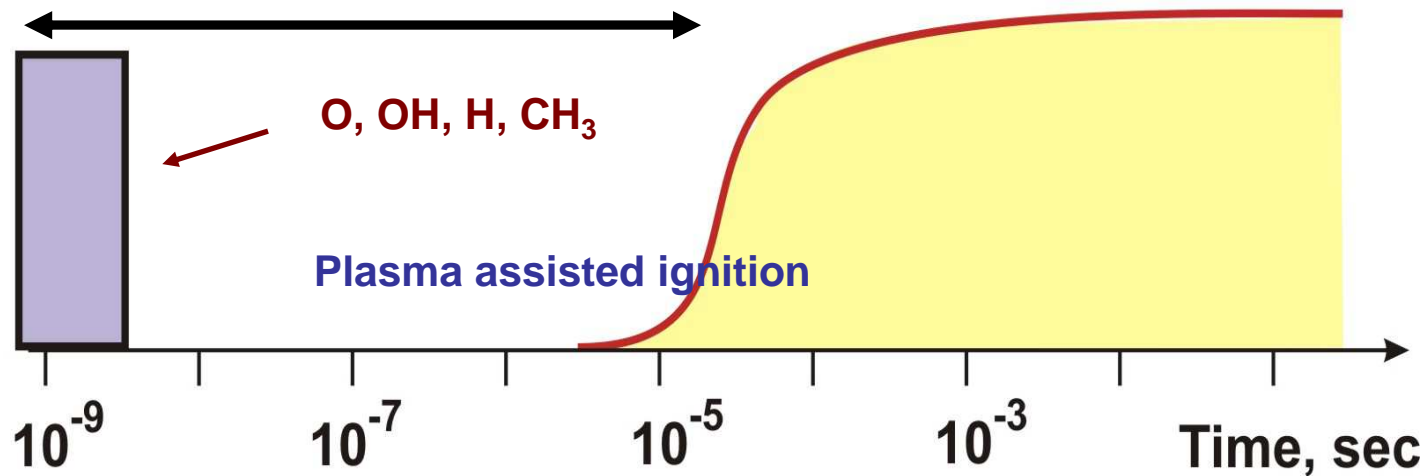
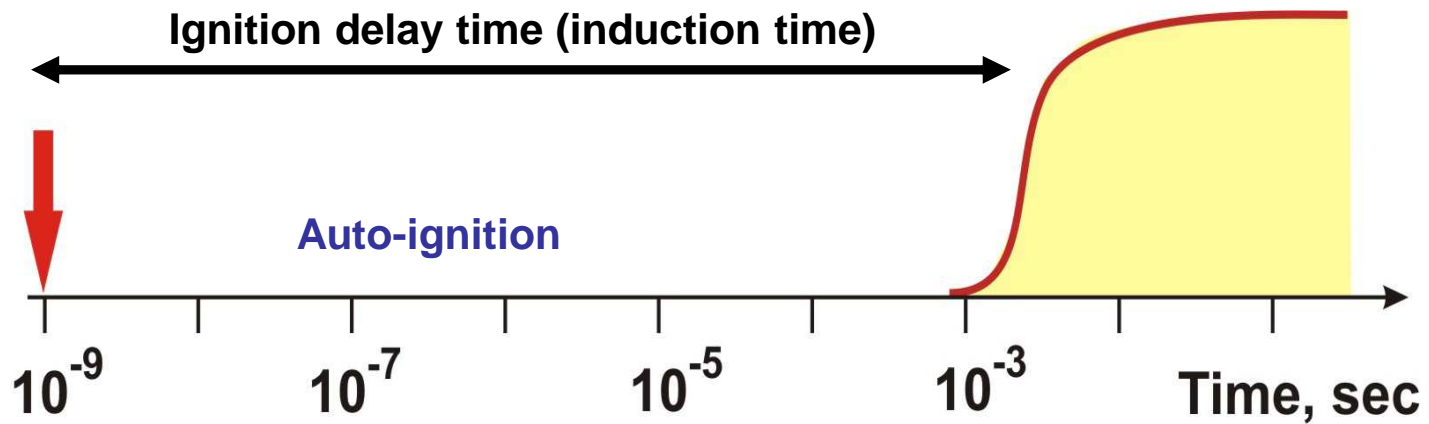


# Dielectric section of a shock tube

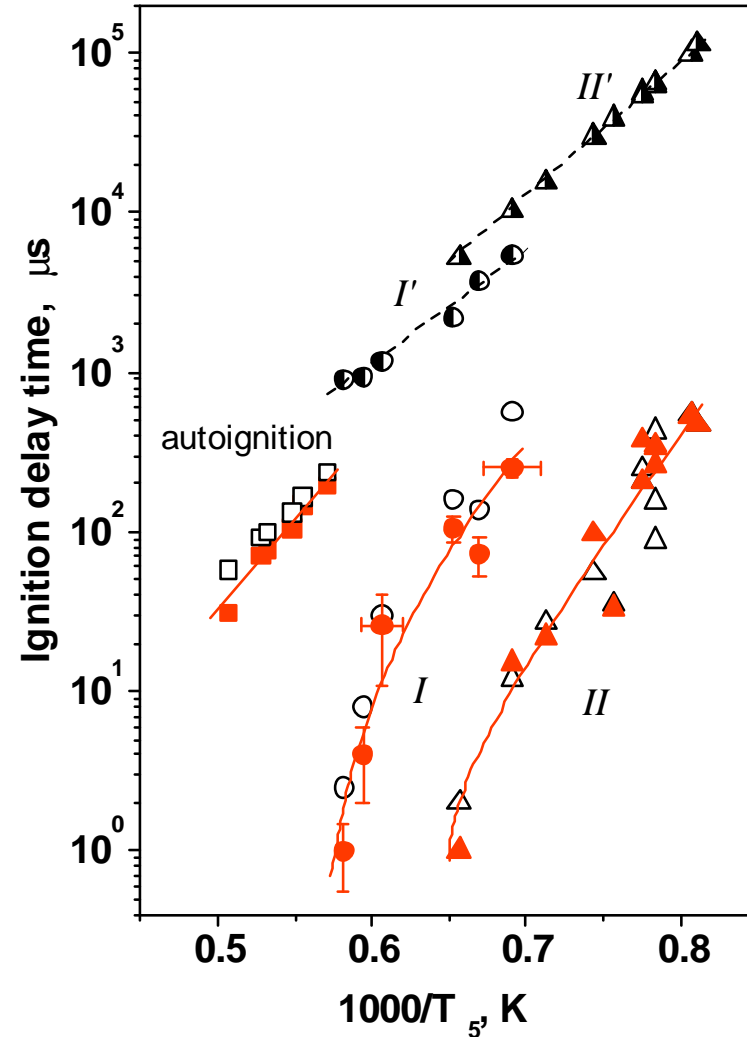
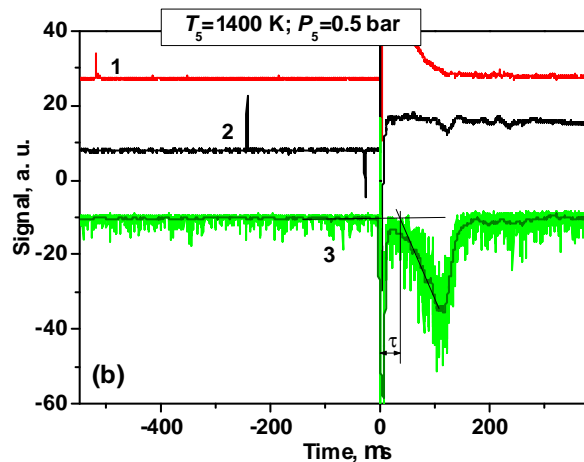
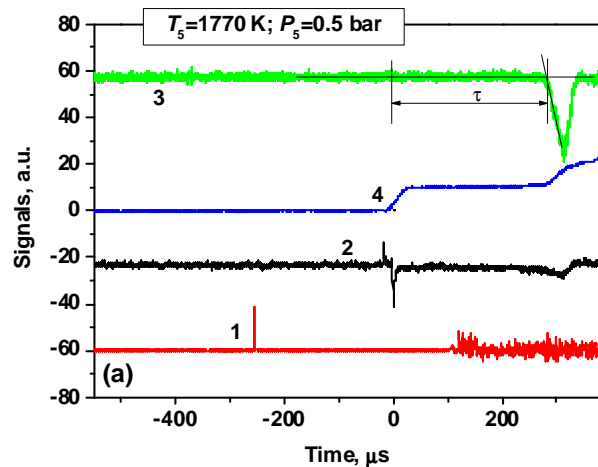


Starikovskaia S M, Kukaev E N, Kuksin A Yu,  
Nudnova M M and Starikovskii  
Comb. and Flame, 2004, 139, 177-87

# The idea of the experiment

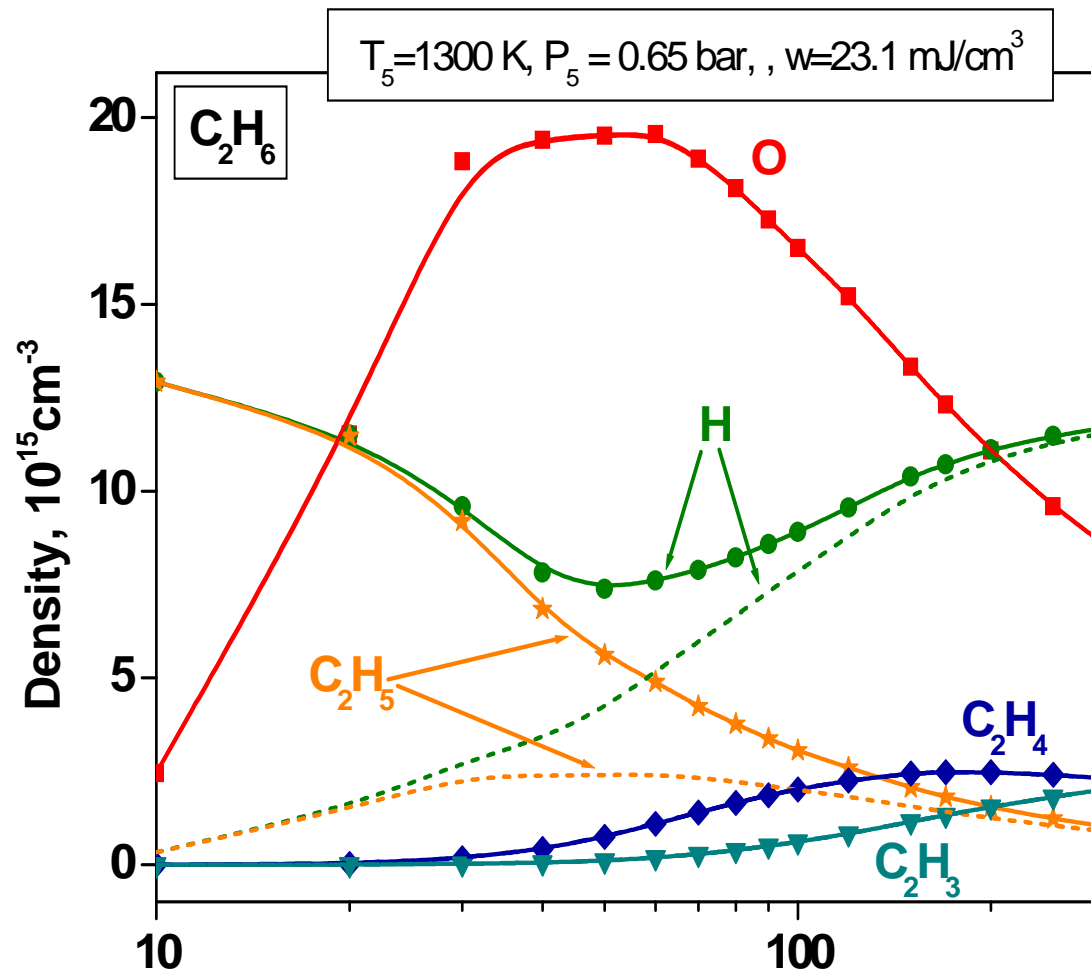


# Shift of the ignition delay time: (CH<sub>4</sub>:O<sub>2</sub>):Ar = 90:10 mixture



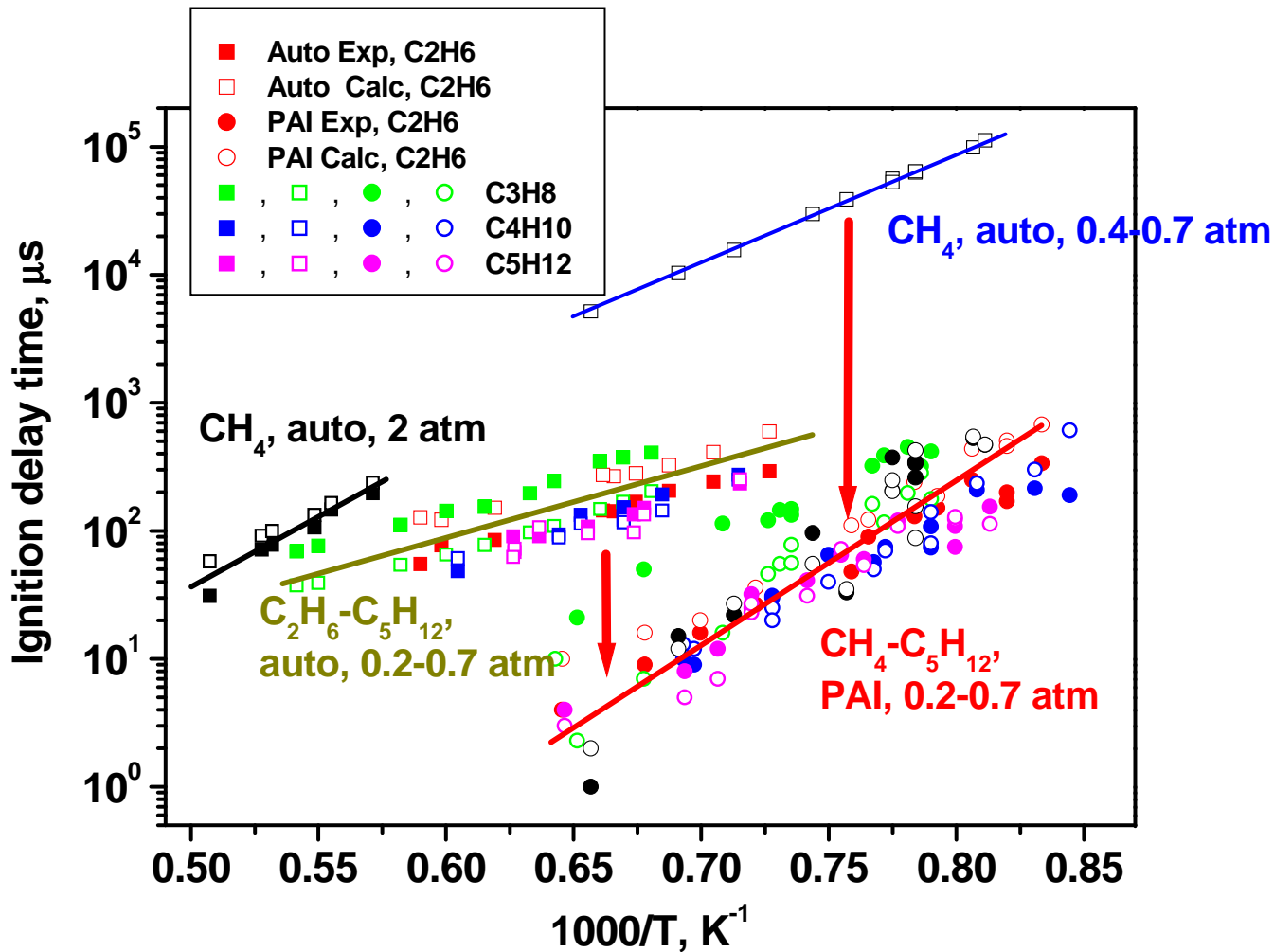
I N Kosarev, N L Aleksandrov, S V Kindysheva,  
S M Starikovskaia, A Yu Starikovskii, *Comb Flame*, 154 (2008) 569-586

# Dissociation in a nanosecond discharge (C<sub>2</sub>H<sub>6</sub>:O<sub>2</sub>):Ar, Hayashi (C<sub>2</sub>H<sub>6</sub>), Braginsky (O<sub>2</sub>), Tachibana (Ar)

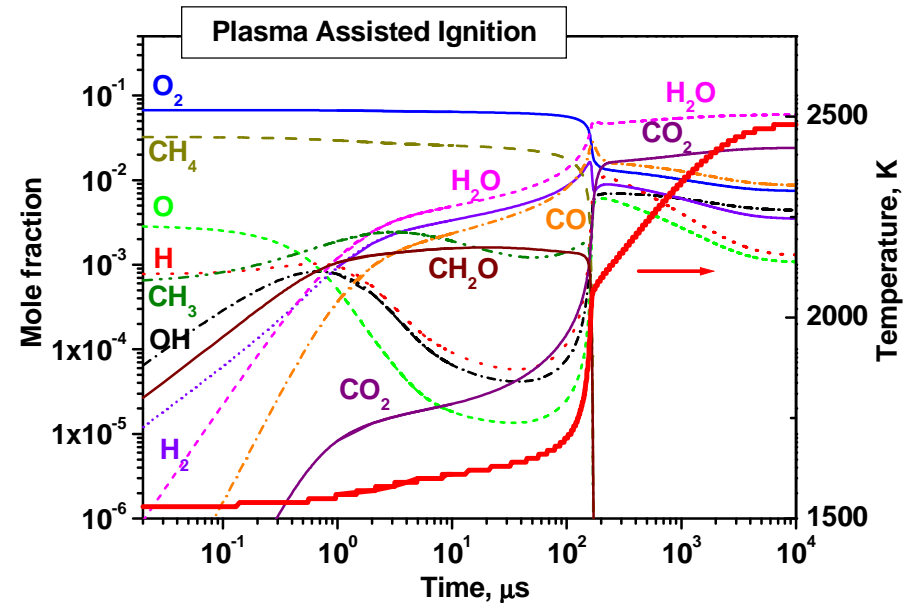
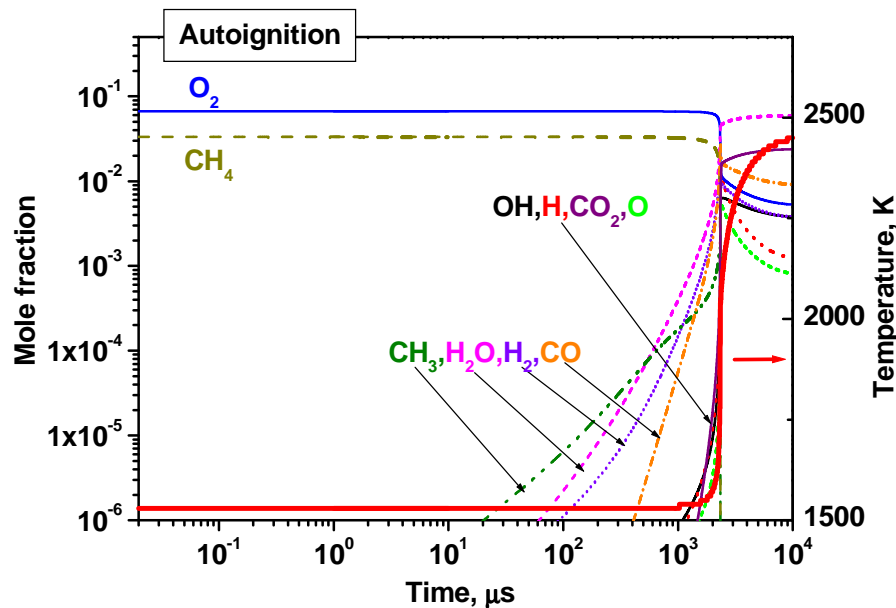


I N Kosarev, N L Aleksandrov, S V Kindysheva, S M Starikovskaia, A Yu Starikovskii, *Comb Flame*, 156 (2009) 221-233

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



# Kinetics of the ignition: kinetic curves ( $T_5=1530$ K, $n_5=5 \times 10^{18}$ cm $^{-3}$ )



Plasma assisted ignition is characterized by:

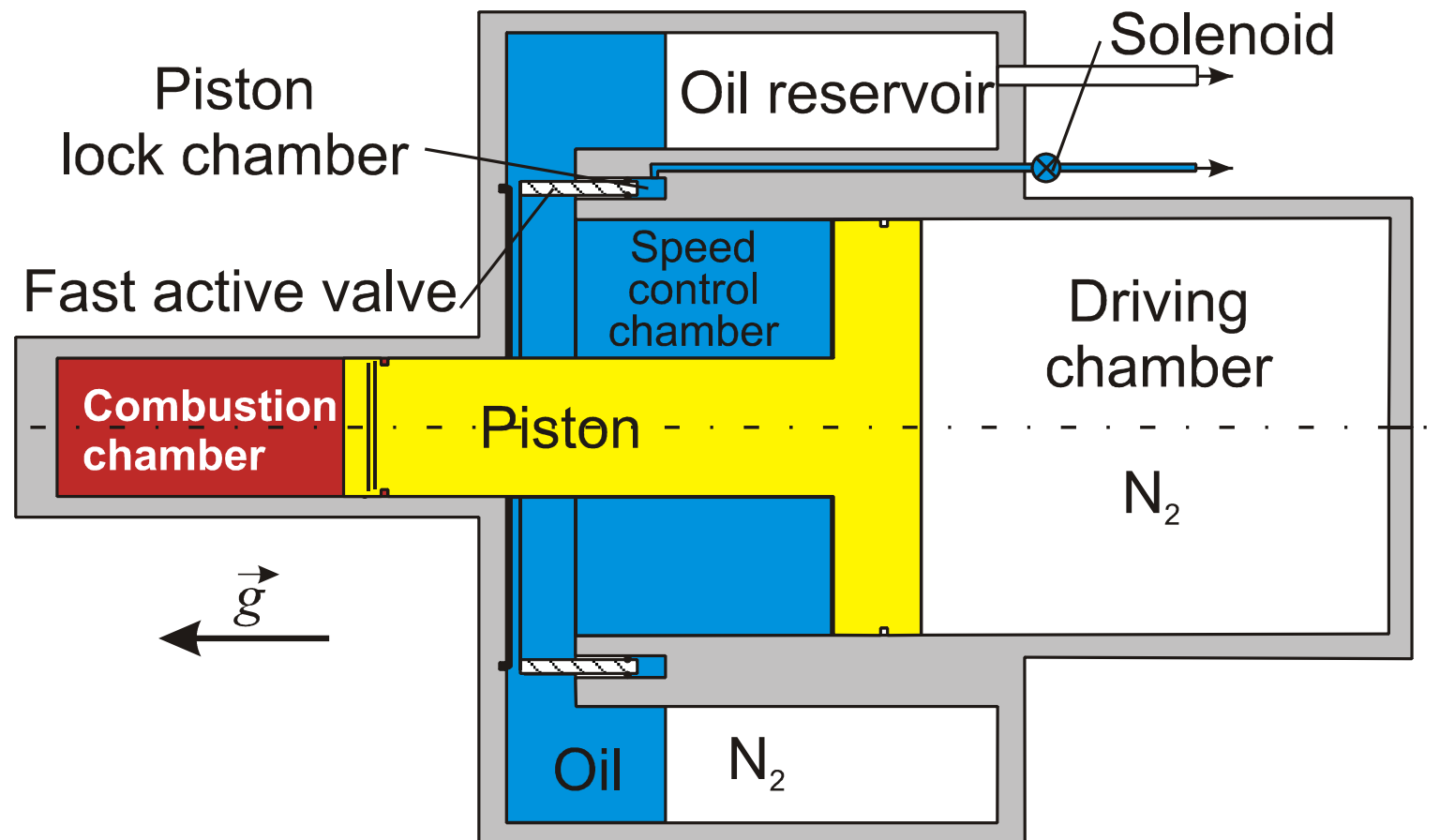
- slow increase of gas temperature
- developed kinetics of intermediates
- partial fuel conversion during induction time

## II. Rapid compression machine (RCM) experiments: high $P$ and [relatively] low $T$

# Rapid compression machine (RCM)



P.Park and J.C.Keck, SAE Paper 900027

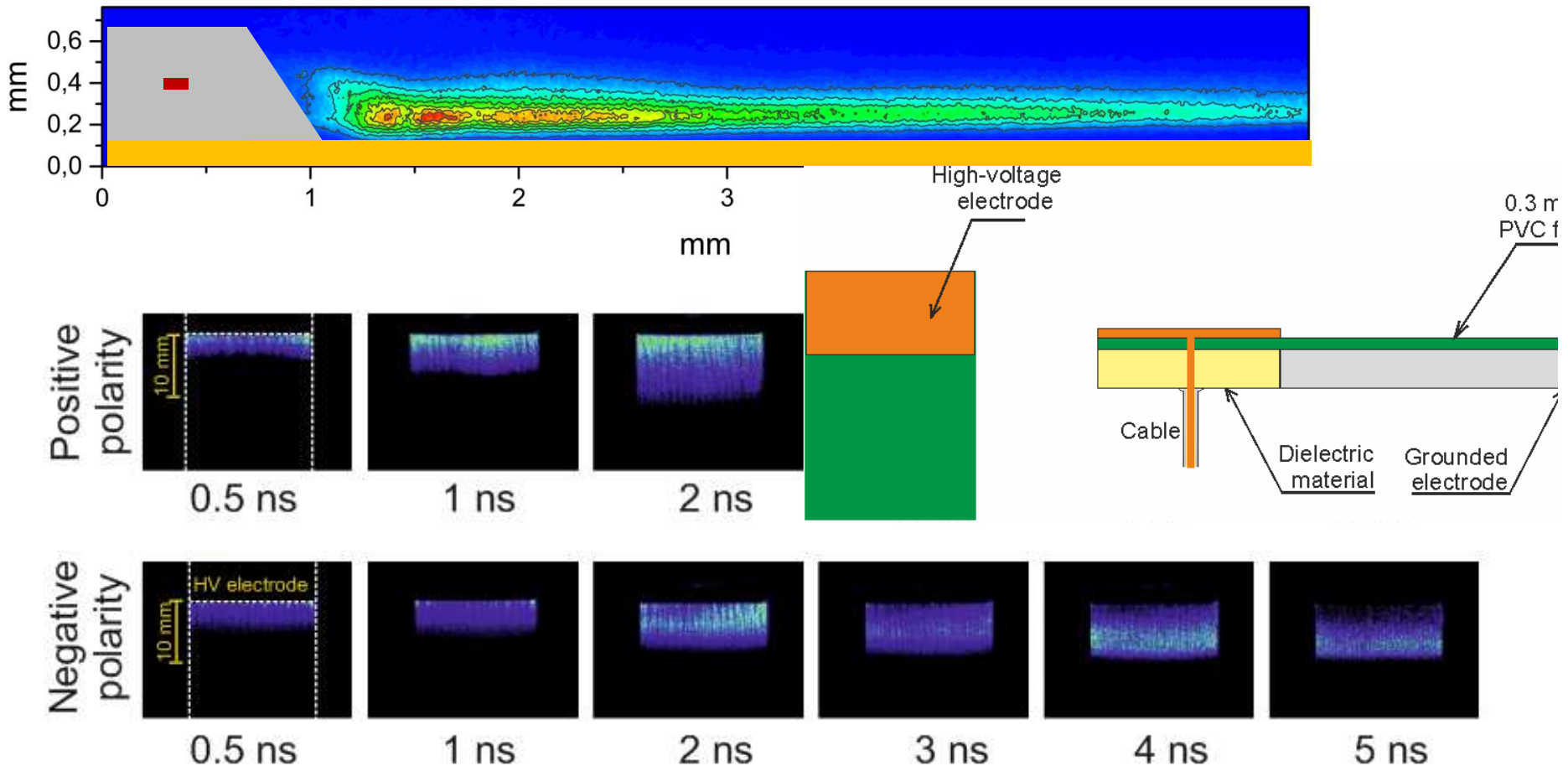




# Nanosecond surface dielectric barrier discharge (nSDBD, for flow control)



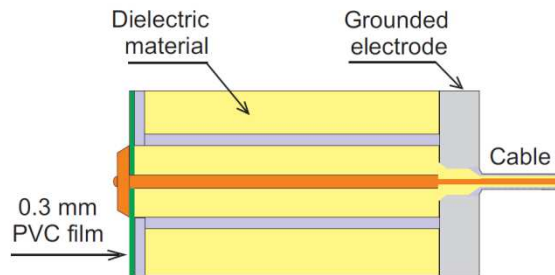
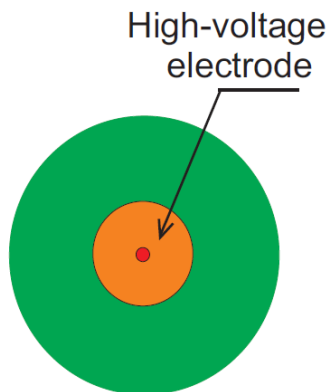
negative 1.5 ns



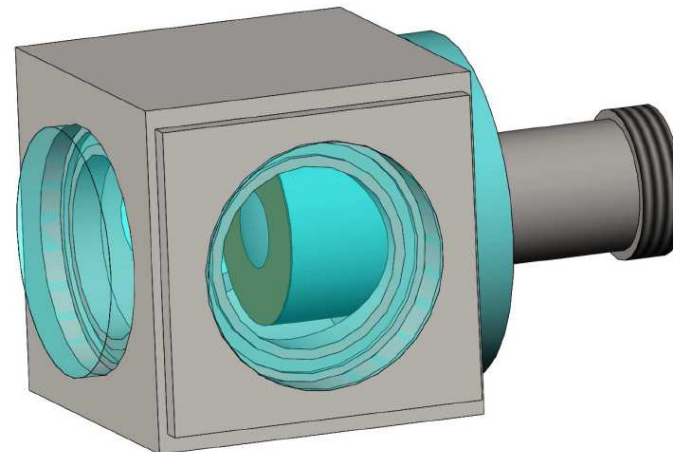
# Cylindrical electrode system (for PAC)



## Electrode configuration

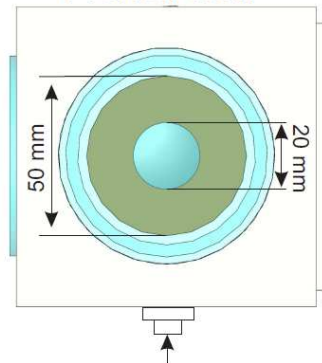


## Cubic HP chamber

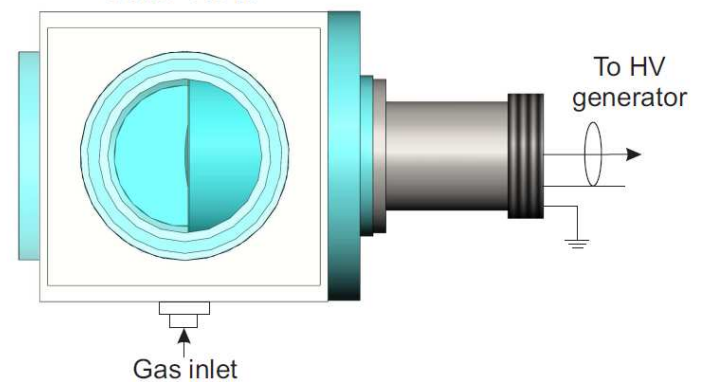


HV electrode,  
2 cm in diameter

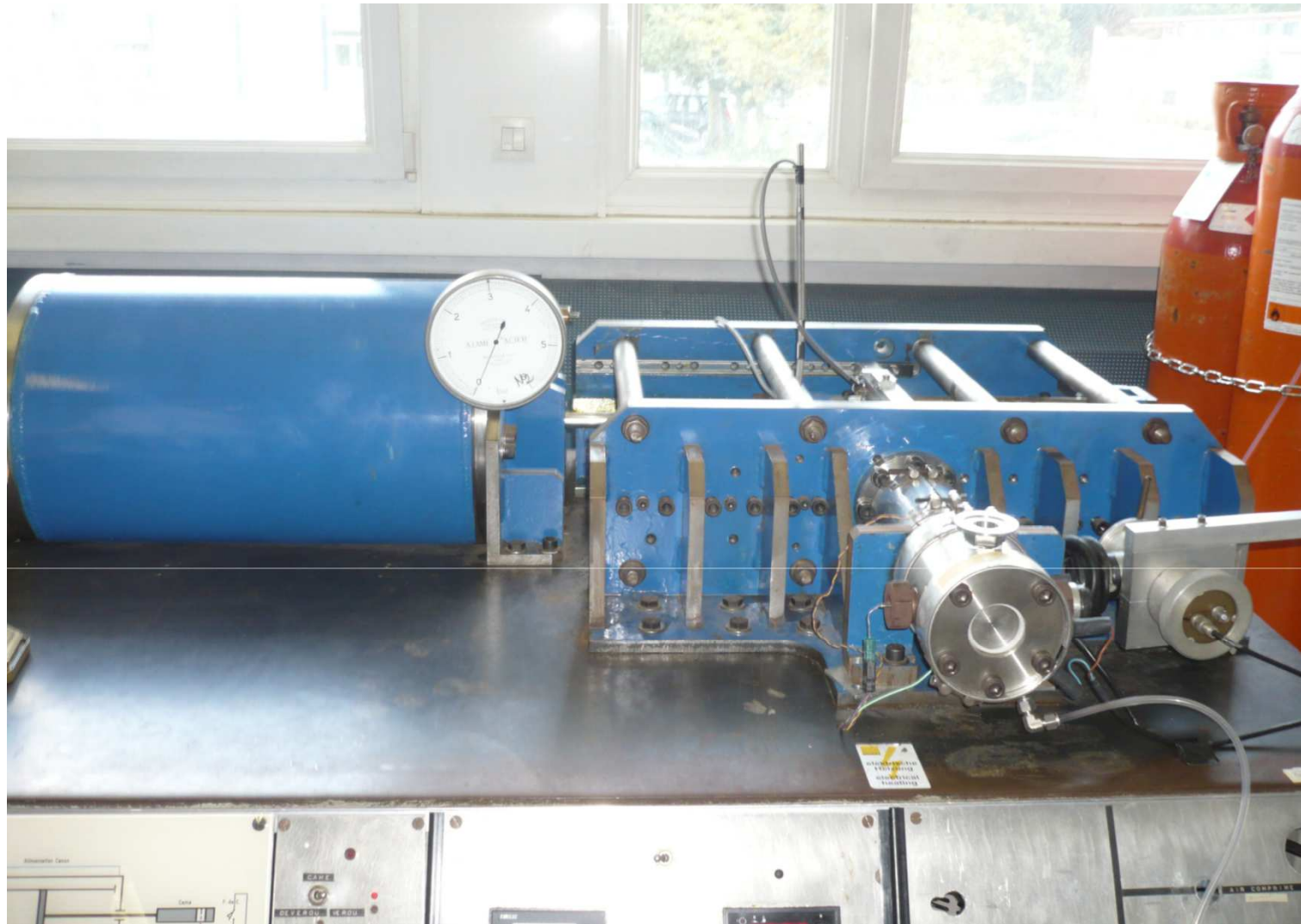
Frontal view



Side view



# Rapid compression machine (RCM), Lille



# Mixtures used in experiments:



1)  $\text{CH}_4/\text{O}_2/\text{Ar}$ ,  $\phi=1, 0.5, 0.3$ , 70-75 % of Ar

2)  $n\text{-C}_4\text{H}_{10}/\text{O}_2/\text{Ar}/\text{N}_2$ ,  $\phi=1$ , 38 % of  $\text{N}_2$ , 38 % of Ar

3)  $n\text{-C}_4\text{H}_{10}/\text{O}_2/\text{Ar}$ ,  $\phi=1$ , 76 % of Ar

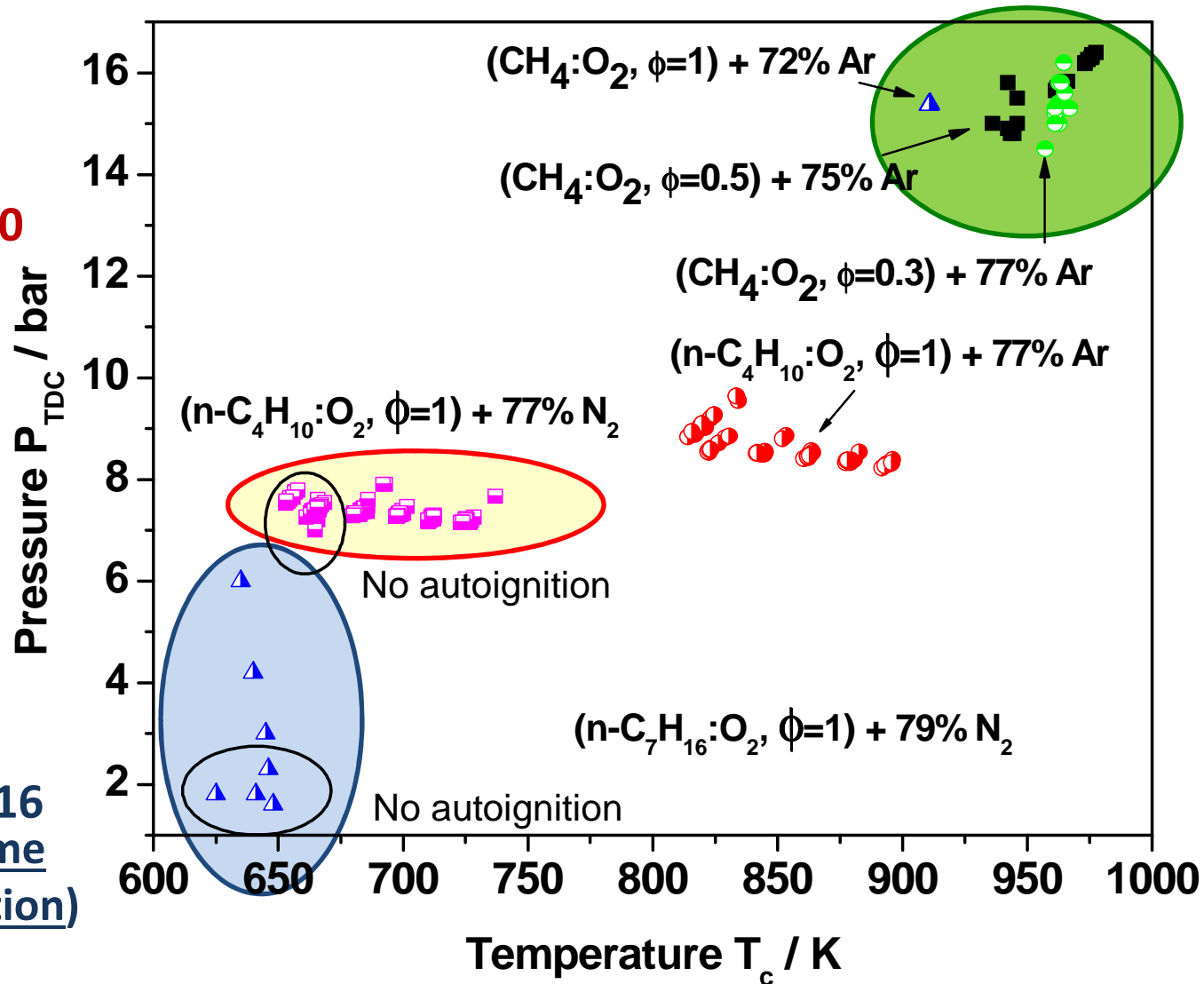
4)  $n\text{-C}_4\text{H}_{10}/\text{O}_2/\text{N}_2$ ,  $\phi=1$ , 76 % of  $\text{N}_2$

# P-T diagram for RCM experiments



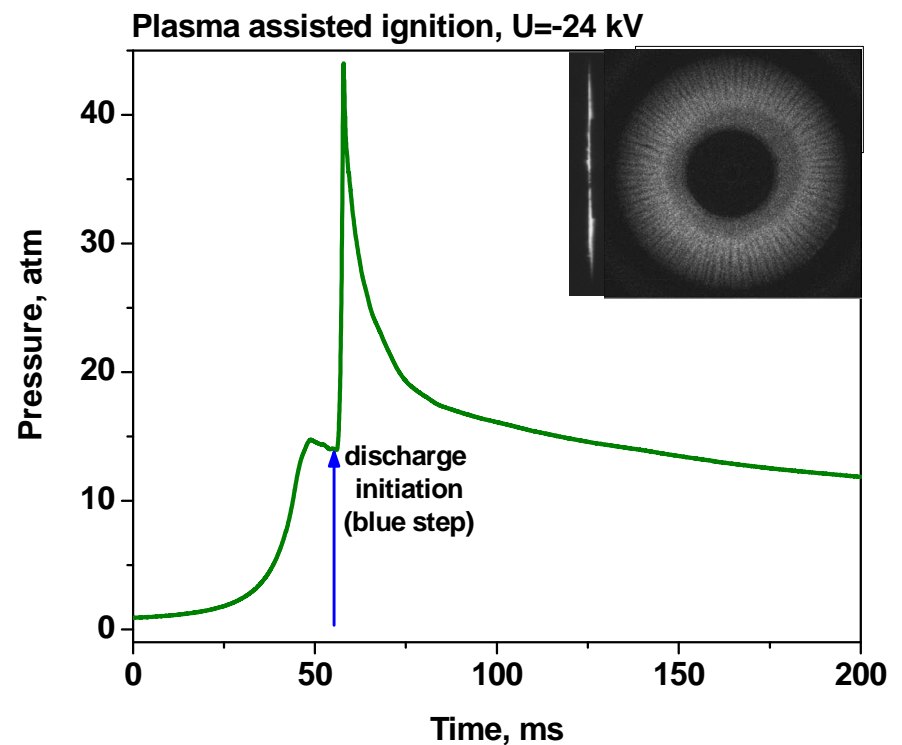
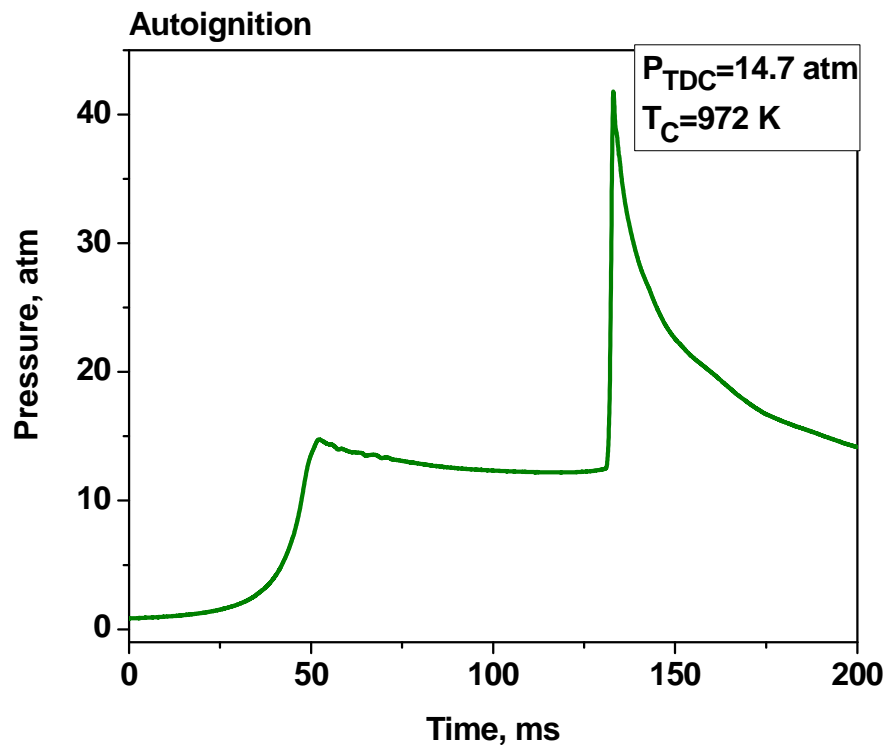
$n\text{-C}_4\text{H}_{10}$

$n\text{-C}_7\text{H}_{16}$   
(cool flame  
modification)



CH<sub>4</sub>

# Autoignition vs plasma ignition in RCM at $P_{TDC}=15$ bar and $T_C=970$ K, $(CH_4:O_2)+76\%Ar$

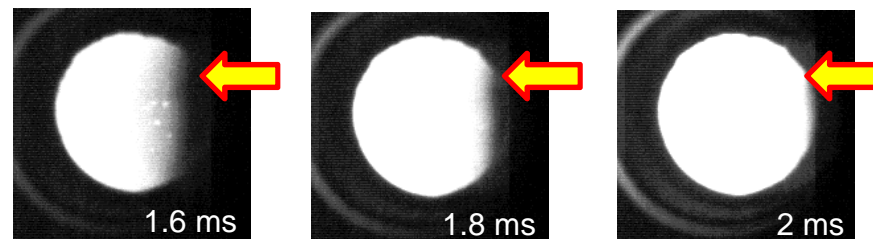
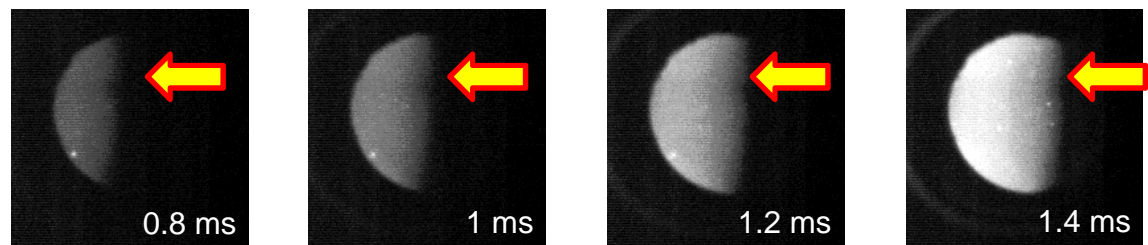
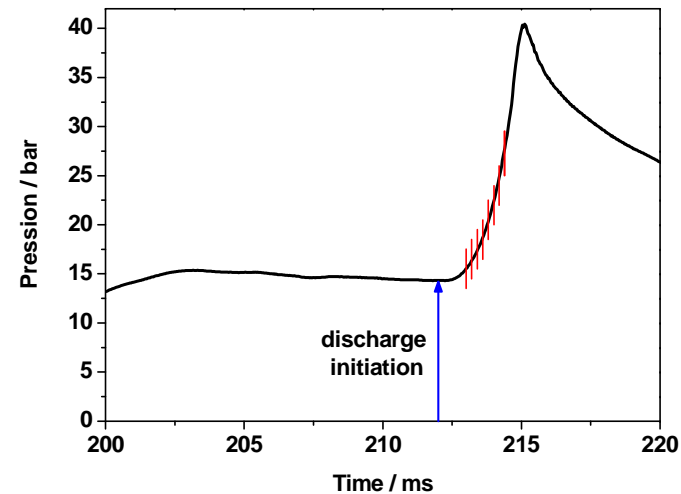
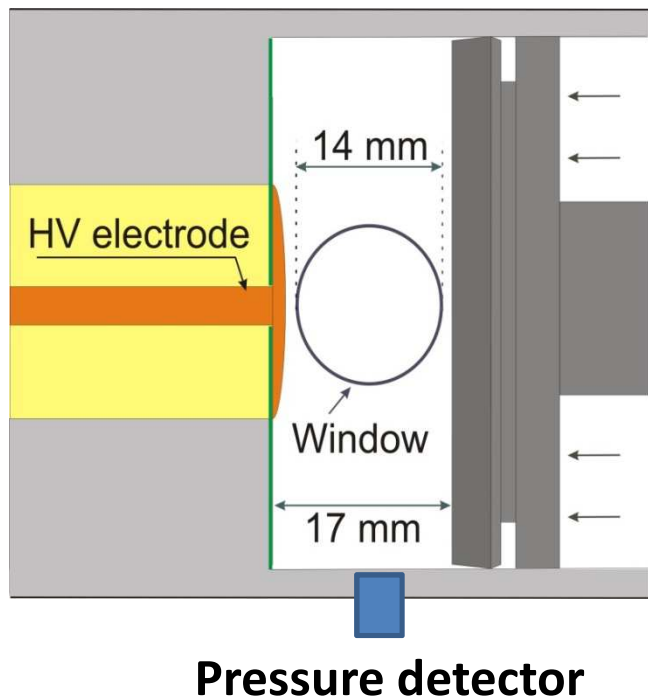




# Pressure trace and corresponding fast imaging of flame propagation

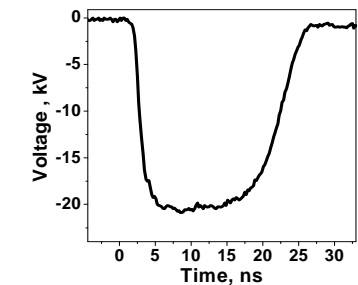
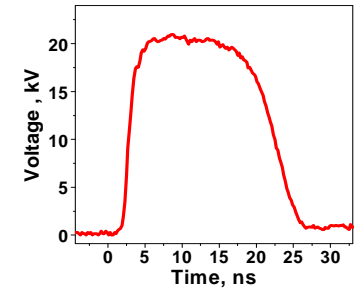
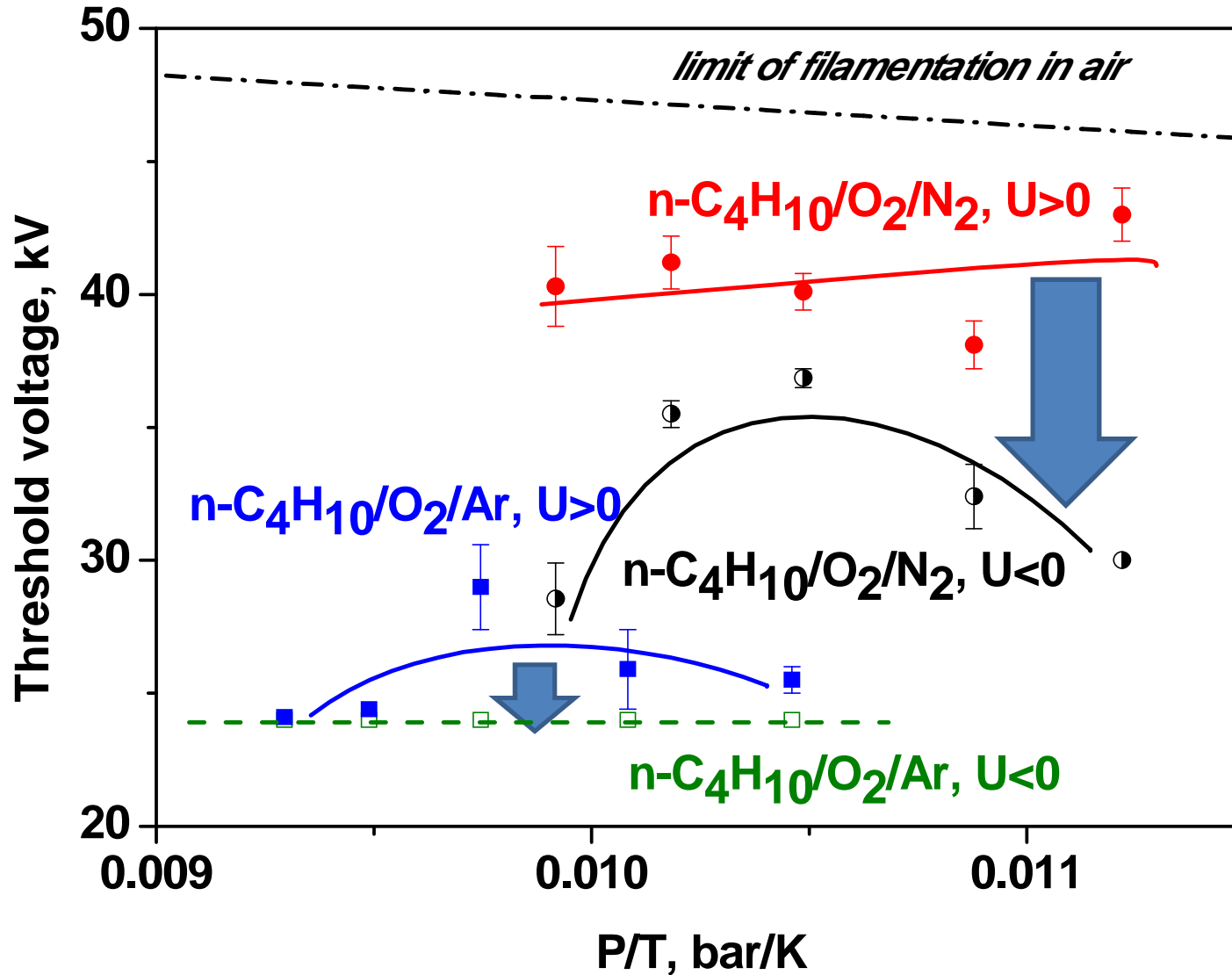


S.A. Stepanyan, M.A. Boumehdi, G. Vanhove, P. Desgroux, S.M. Starikovskaia, N.A. Popov,  
 Comb. Flame, 162 (2015) 1336-1349



$\text{CH}_4:\text{O}_2$ , ER=1 + 70% Ar,  
 $T_C=947$  K,  $P_{TDC}=15.4$  bar

# Ignition threshold and polarity of the high-voltage pulse



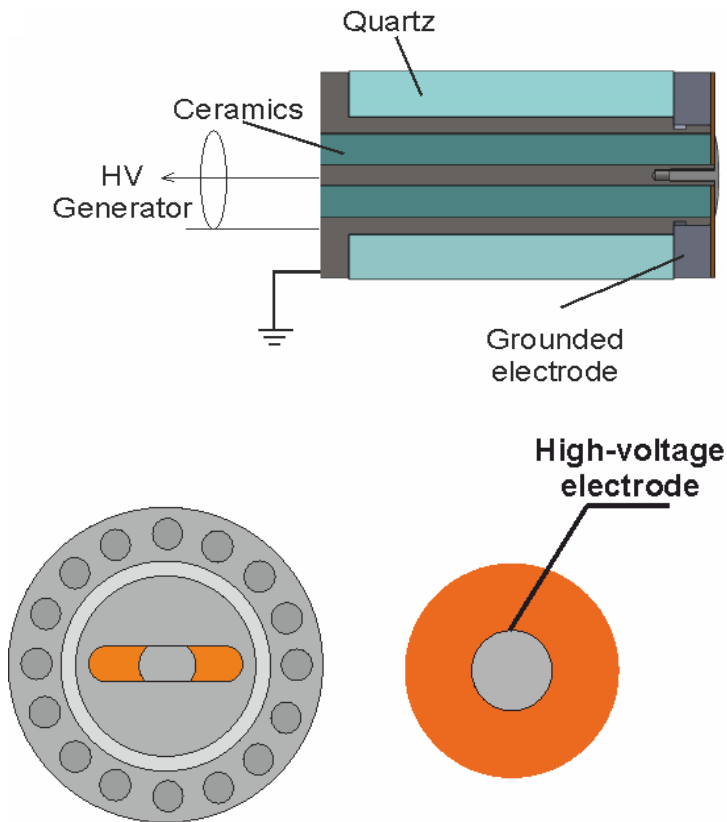


III. High pressure high  
temperature (HPHT) chamber  
experiments:  
high P and low T [ $T=300$  K]

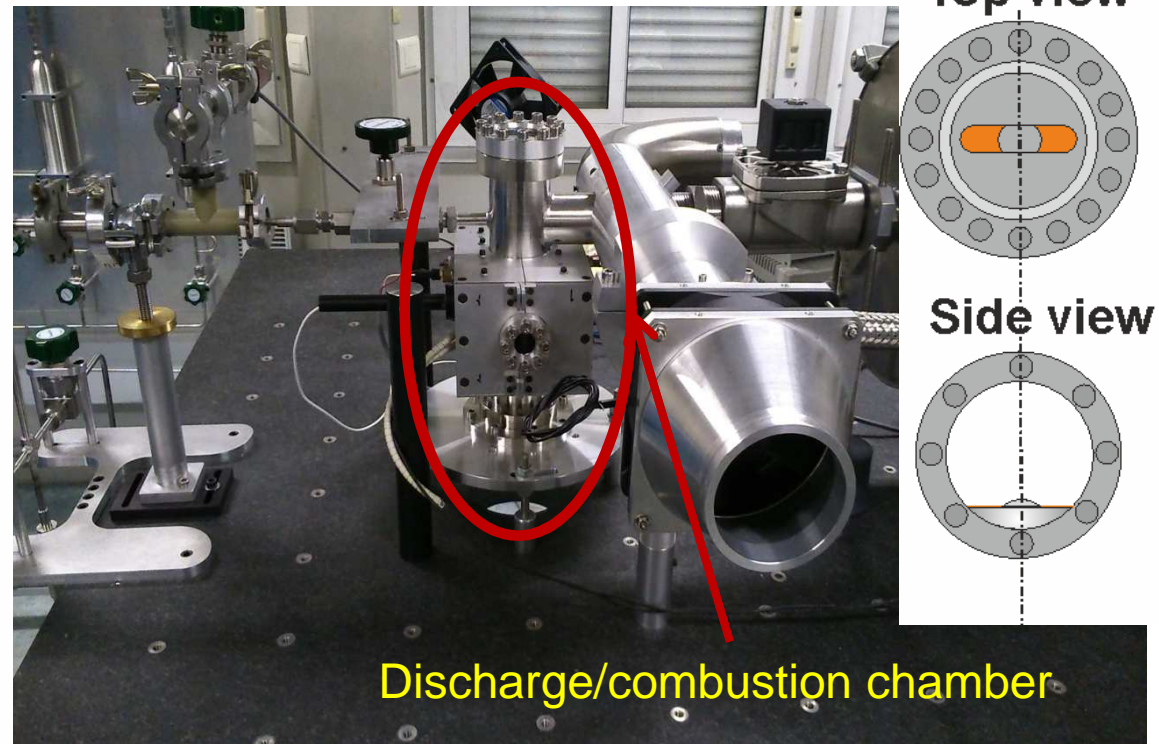
# High-Pressure and High-Temperature (HPHT) discharge/combustion chamber



## Electrode configuration



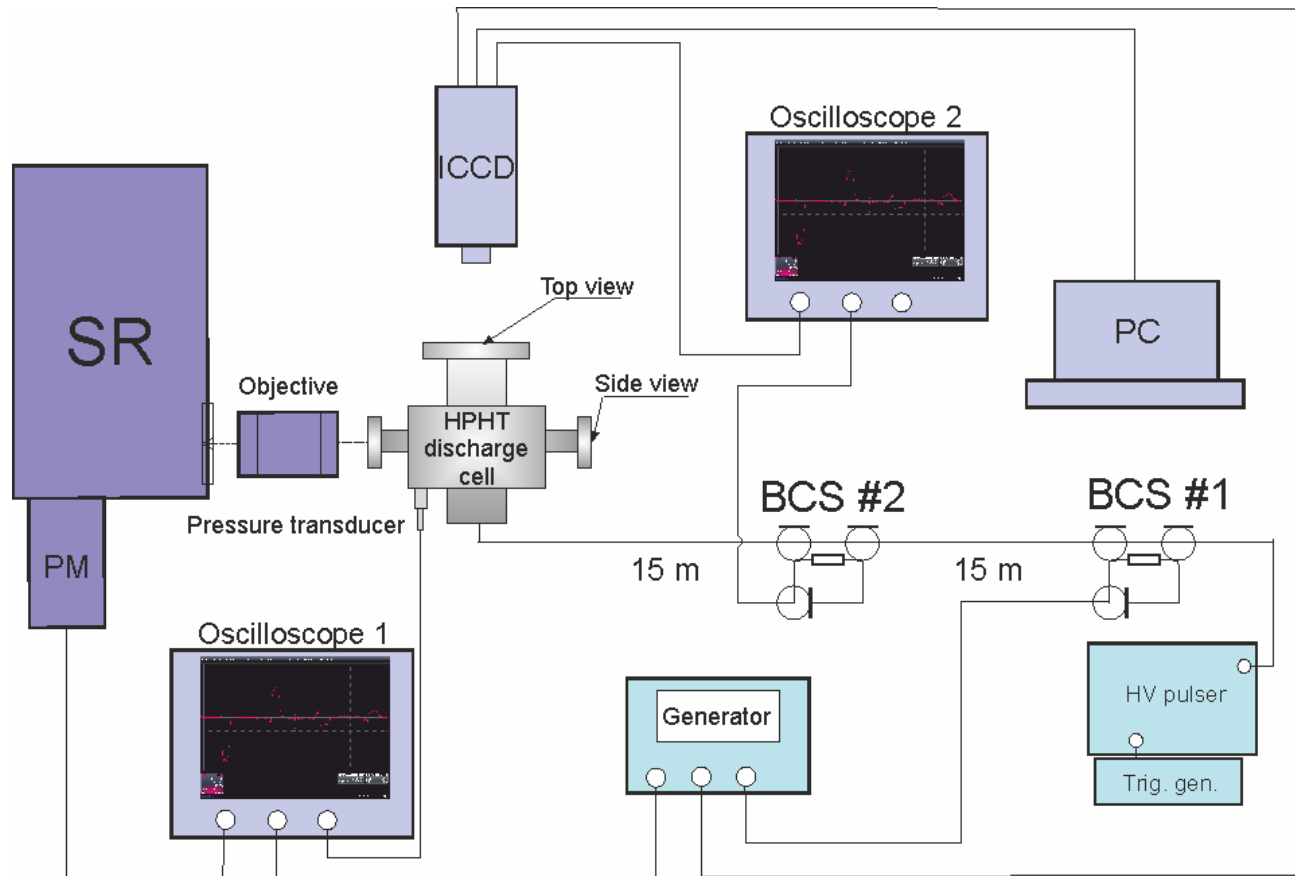
## General view of the HPHT setup



# Experimental setup



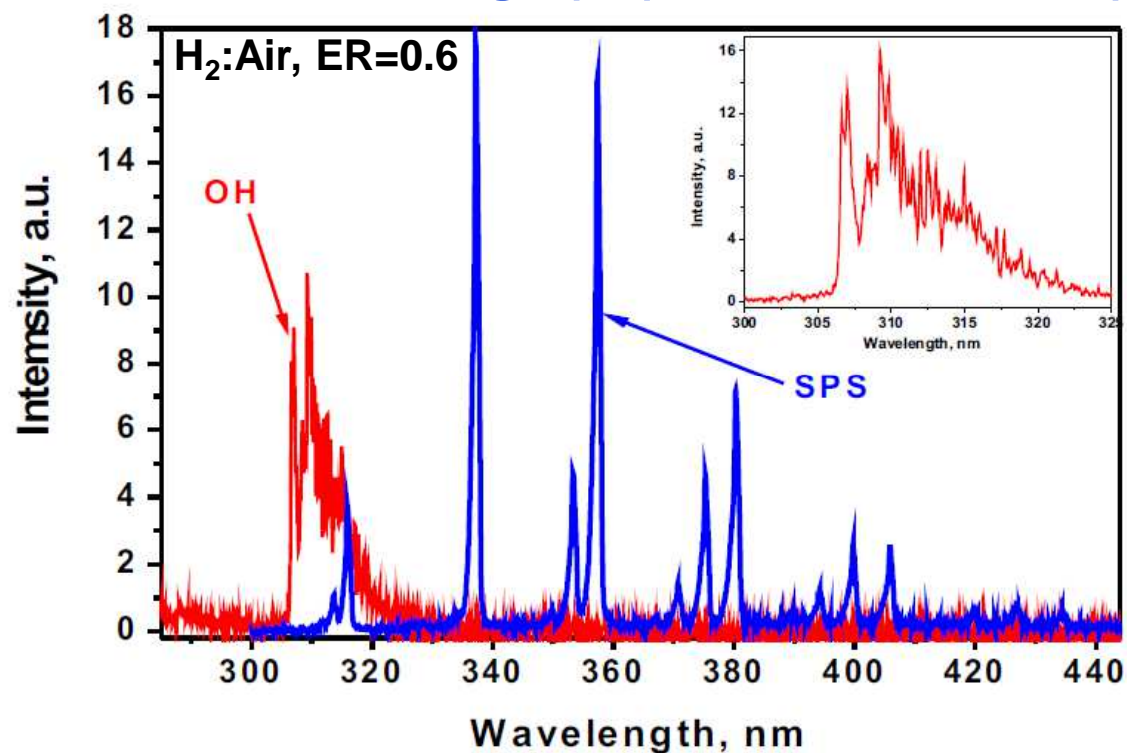
The scheme of experimental setup. SR – Spectrograph, ICCD – camera, PC – computer, BCS – back current shunt,



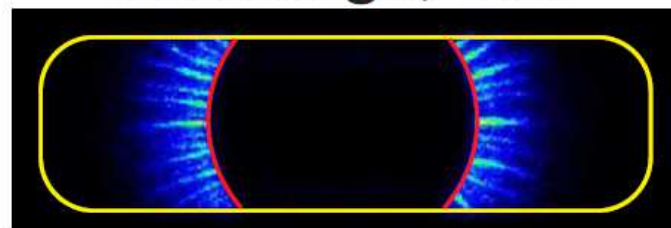
# nSDBD and Flame Initiation, P=3 bar, U=-50 kV



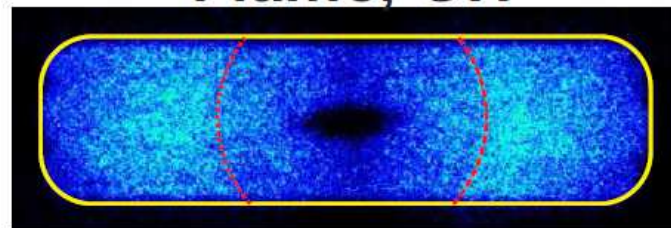
## Discharge (ns) and combustion (ms) emission patterns



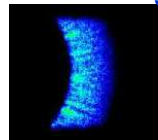
Discharge, SPS



Flame, OH

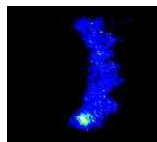


ns discharge



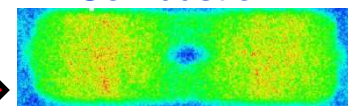
$N_2(v)$ ,  $N_2(A,B,C,\dots)$ , H,  
 $O(^1D)$ ,  $O_2(a^1\Delta_g)$ , etc.

Intermediate chemistry



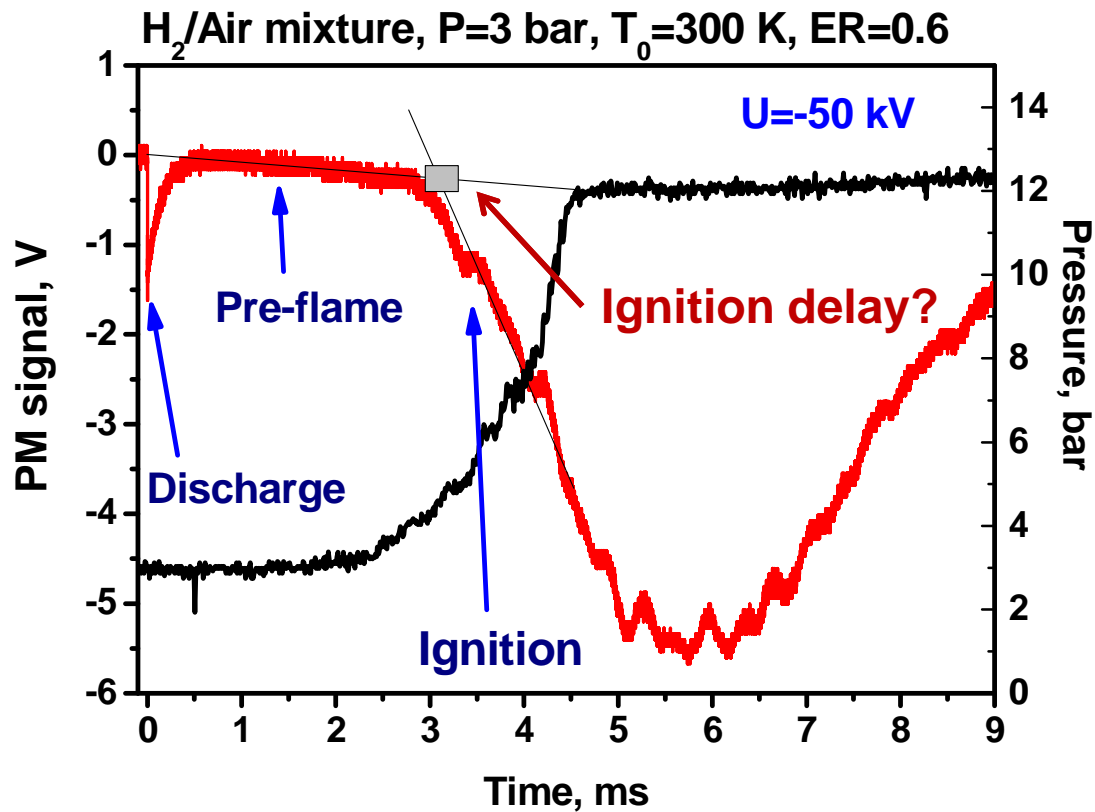
O, OH, H, HO<sub>2</sub>, H<sub>2</sub>O<sub>2</sub>,  
H<sub>2</sub>O, etc.

Flame propagation,  
Combustion



time

# Initiation of Combustion with nSDBD

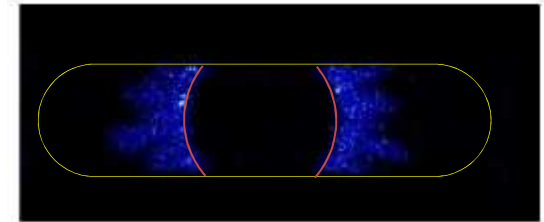


- Yes, for combustion!
- No, for plasma chemistry

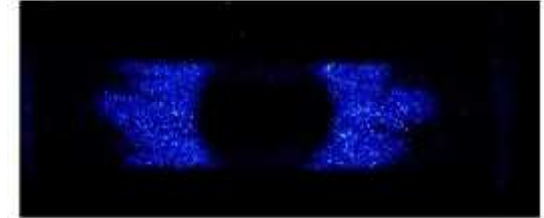
Delay:

ICCD gate: 50  $\mu$ s

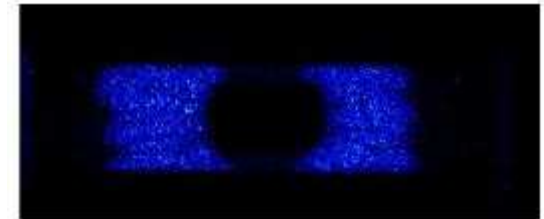
50  $\mu$ s



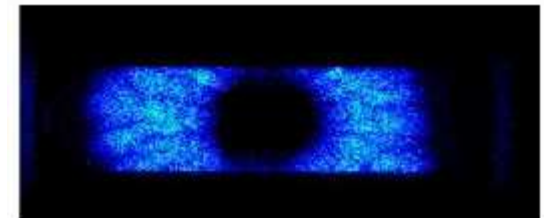
100  $\mu$ s



200  $\mu$ s



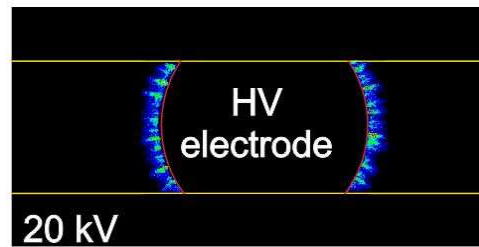
400  $\mu$ s



# Flame Initiation in H<sub>2</sub>/Air ER=0.5, P=6 bar

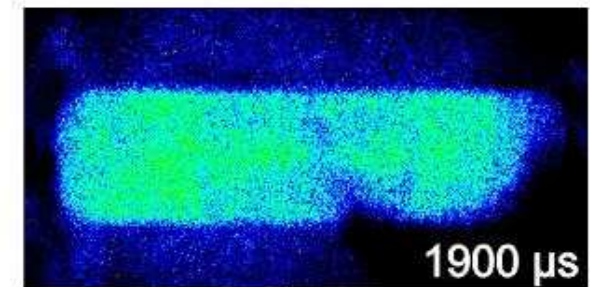
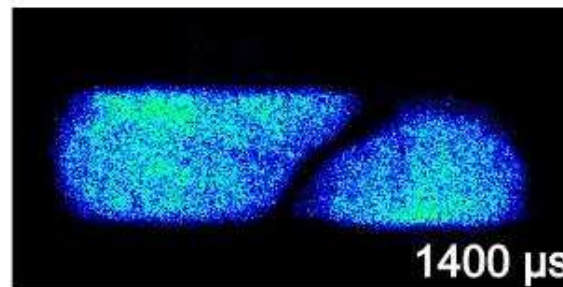
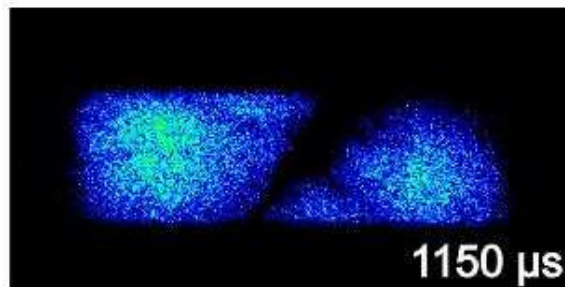
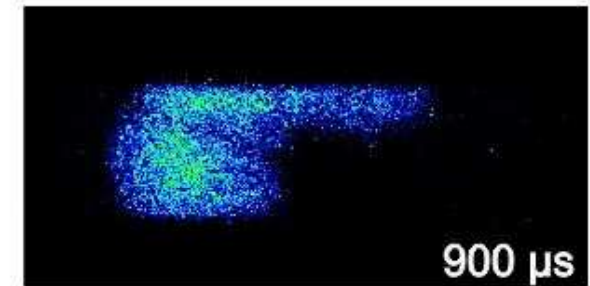
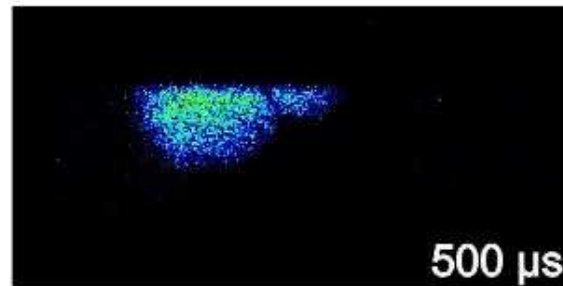
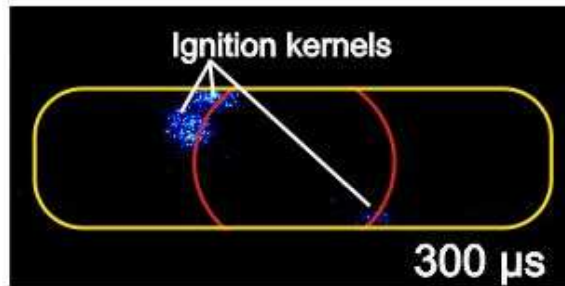


First regime of ignition:  
**Ignition kernels**



**Polarity: U>0**

**Energy deposition  
W= 3 mJ**



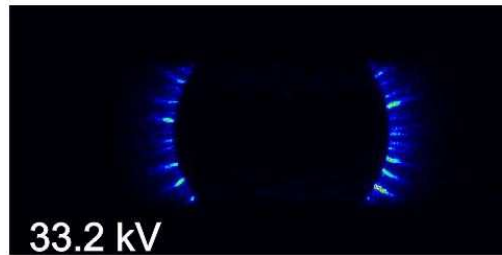
**Ignition with a few ignition kernels near HV electrode. Streamer discharge.  
Pressure 6 bar, Temperature 300 K.**



# Flame Initiation in H<sub>2</sub>/Air ER=0.5, P=6 bar

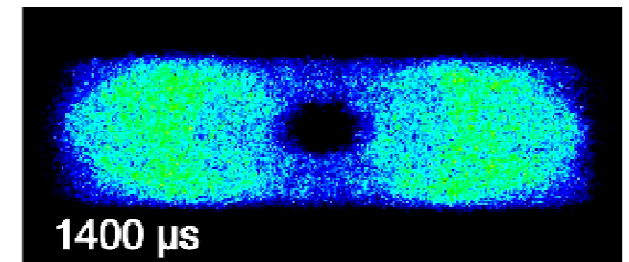
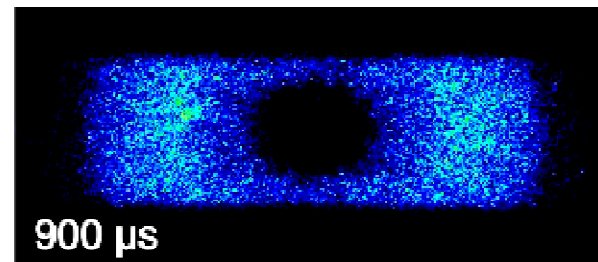
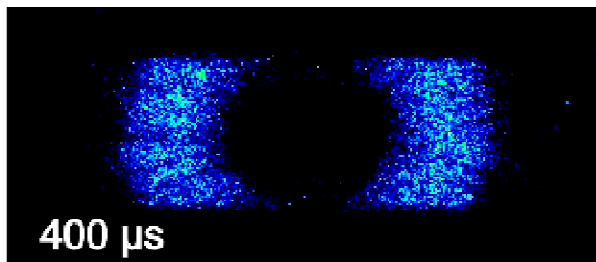
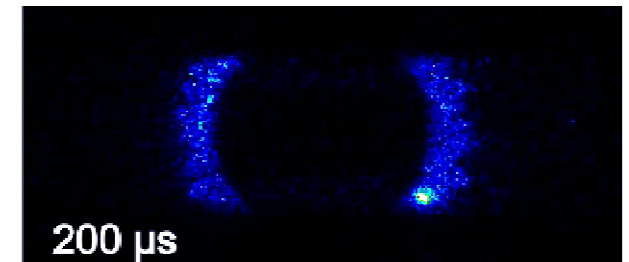
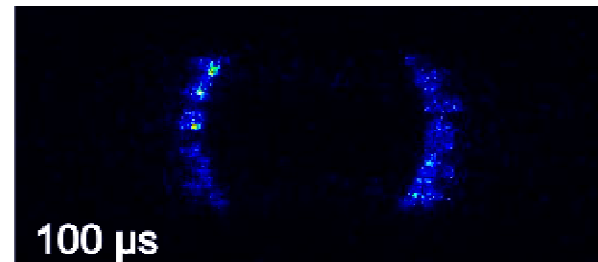
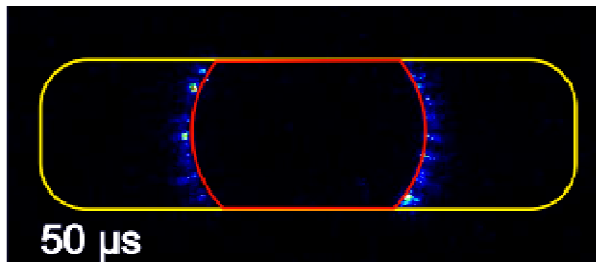


Second regime of ignition:  
Ignition along the  
perimeter of HV electrode



**Polarity: U>0**

**Energy deposition  
W= 4.8 mJ**

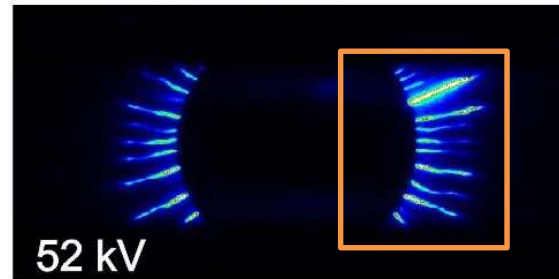


Quasiuniform ignition around HV electrode. Streamer discharge.  
Pressure 6 bar, Temperature 300 K.

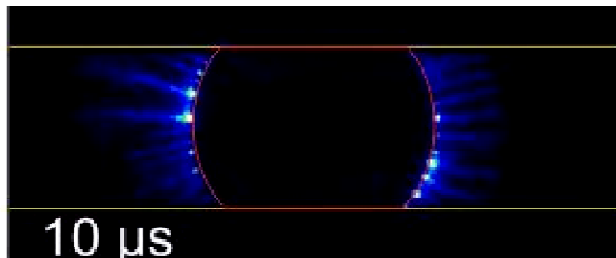
# Flame Initiation in H<sub>2</sub>/Air ER=0.5, P=6 bar



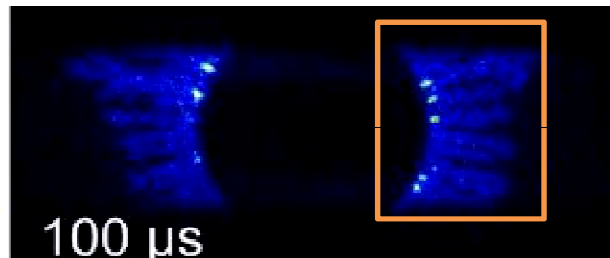
Third regime of ignition:  
**Ignition along the discharge channels**



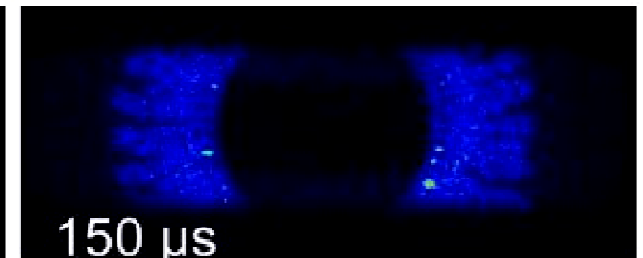
**Polarity: U>0**  
**Energy deposition**  
**W= 12 mJ**



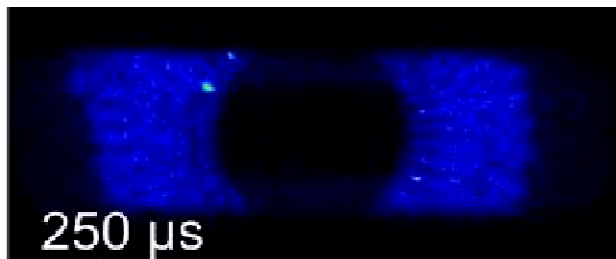
10 μs



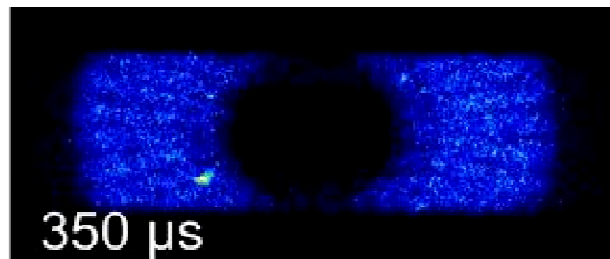
100 μs



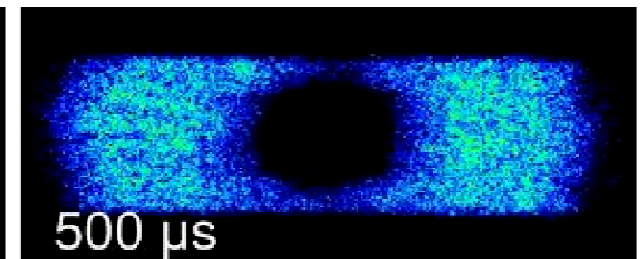
150 μs



250 μs



350 μs



500 μs

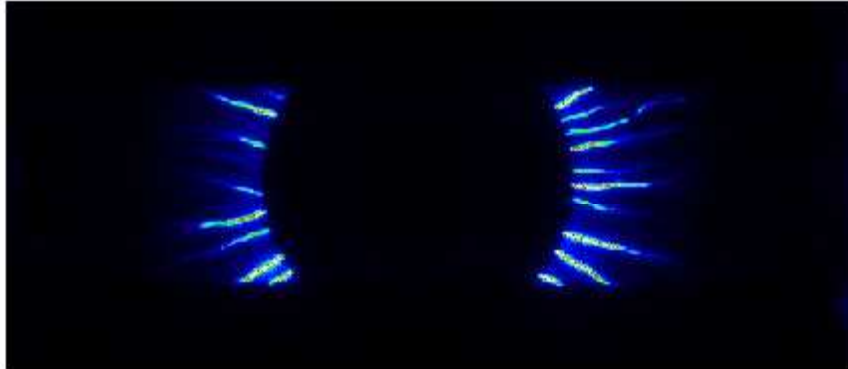
**Ignition along the channels. Filamentary discharge.**  
**Pressure 6 bar, Temperature 300 K.**



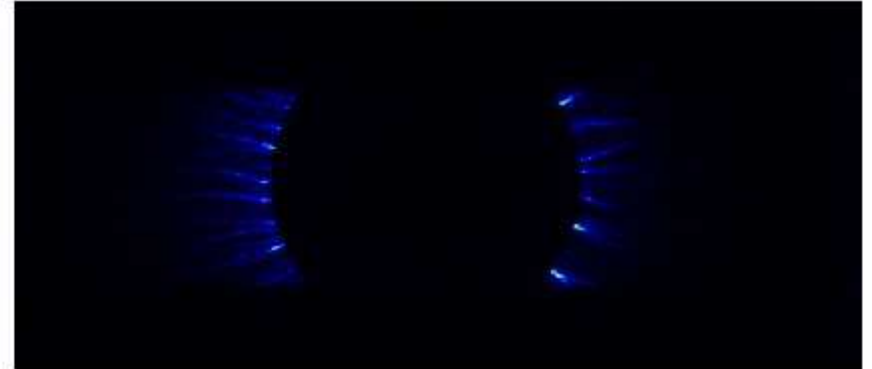
# Discharges in different gas mixtures



Discharge in  
methane/O<sub>2</sub>/Ar



Discharge in  
n-heptane/O<sub>2</sub>/Ar



Discharges in CH<sub>4</sub>/O<sub>2</sub>/Ar (ER=0.6) and  
n-C<sub>7</sub>H<sub>16</sub>/O<sub>2</sub>/Ar (ER=1) mixtures,  
P<sub>0</sub>=3 bar, T<sub>0</sub>=300 K, voltage on the electrode U=+38 kV

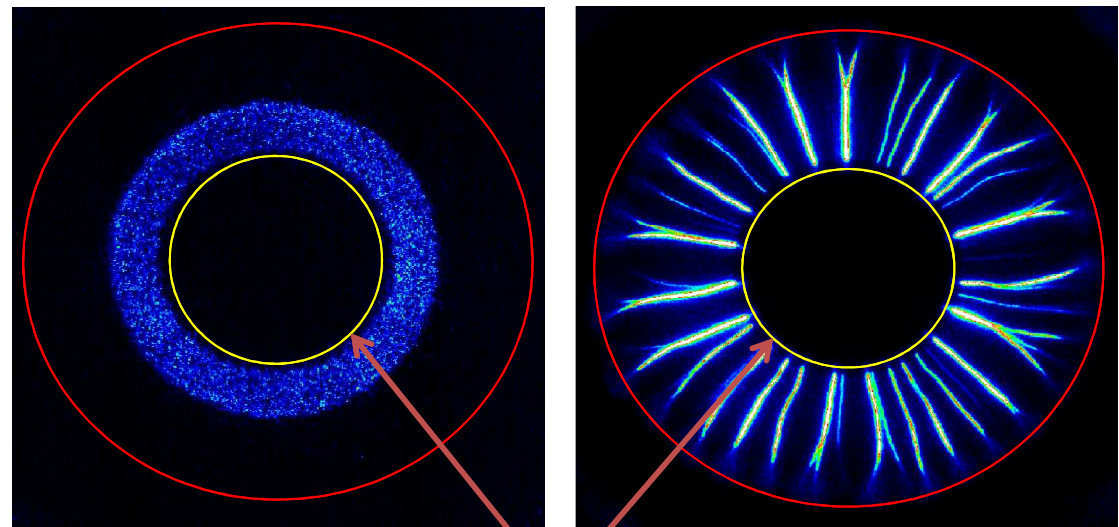
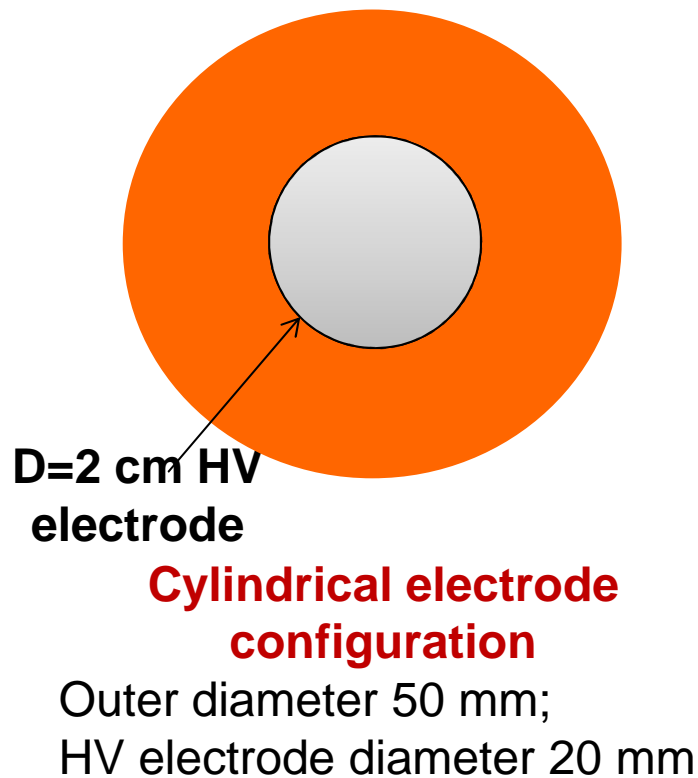
# IV. High pressure surface DBD discharge: streamer-to-filament transition

# Two modes of nSDBD (velocity is a few mm/ns)



Electrode system

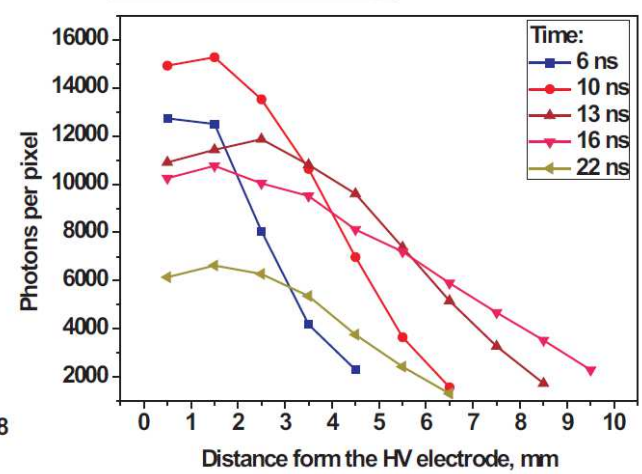
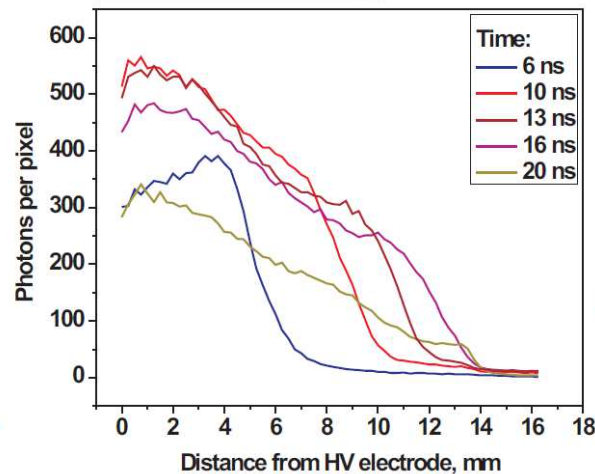
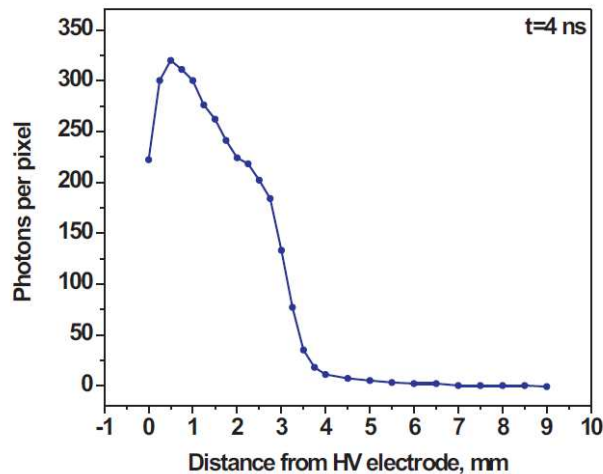
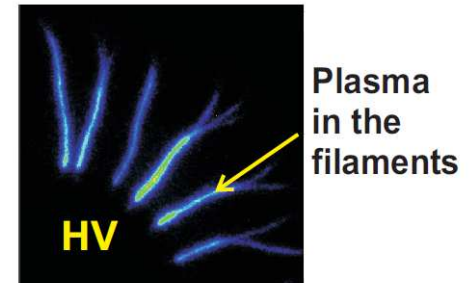
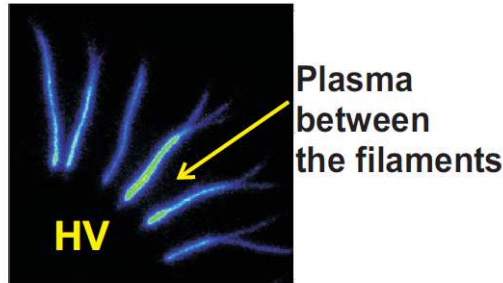
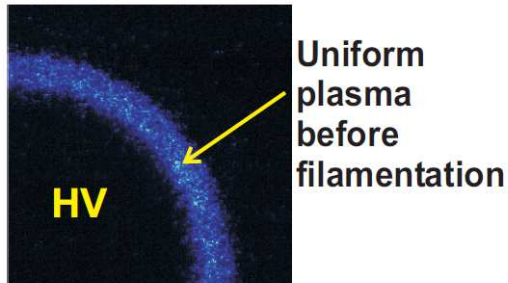
Filamentary mode,  
 $V=-46$  kV, 4 bar, Air



HV electrode

Camera gate is 0.5 ns

# Analysis of emission intensity (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;  $P=4$  bar,  $U=-47$  kV.

# Spatial distribution of emission



	No filter	Filter (340±5) nm	Filter (532±5) nm
Streamers			
Filaments			

# How to get the filament spectra

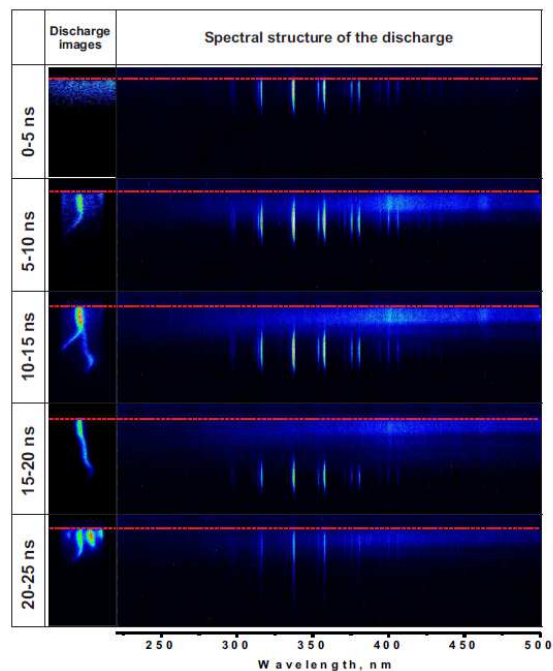
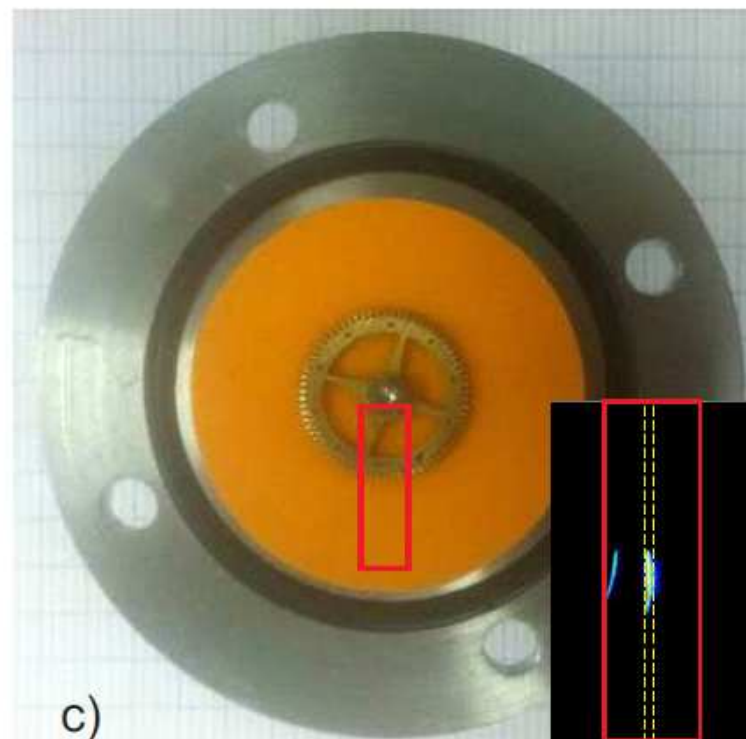
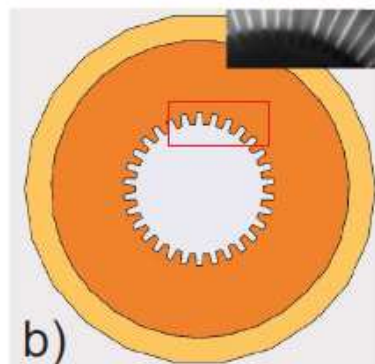
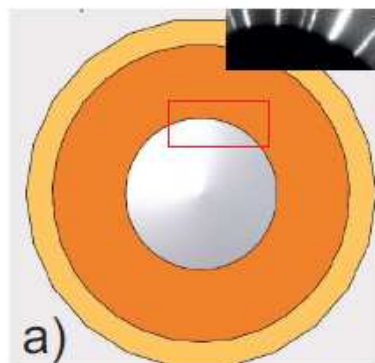
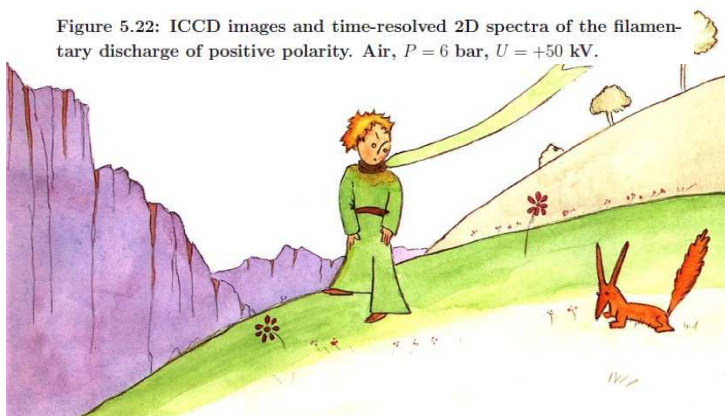
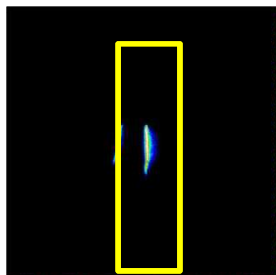
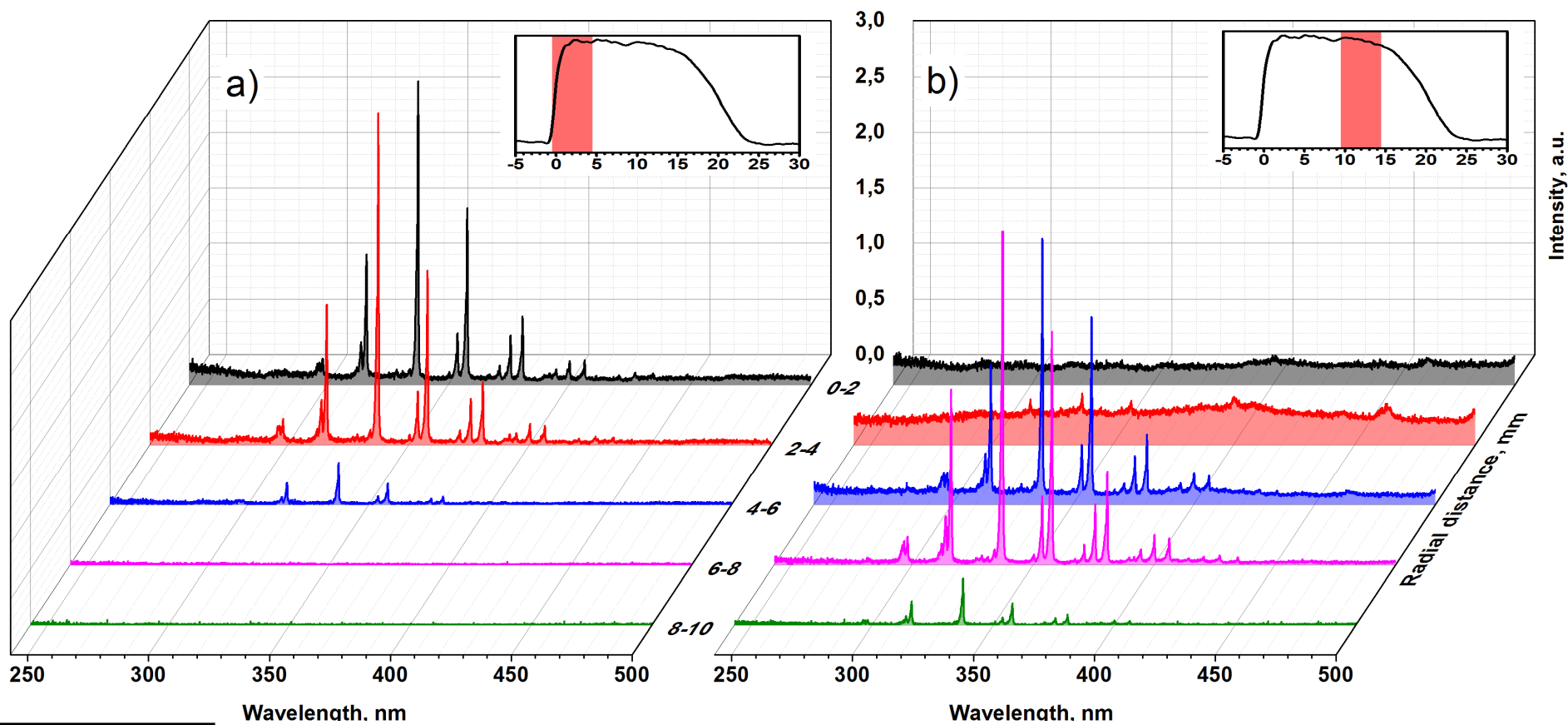


Figure 5.22: ICCD images and time-resolved 2D spectra of the filamentary discharge of positive polarity. Air,  $P = 6$  bar,  $U = +50$  kV.



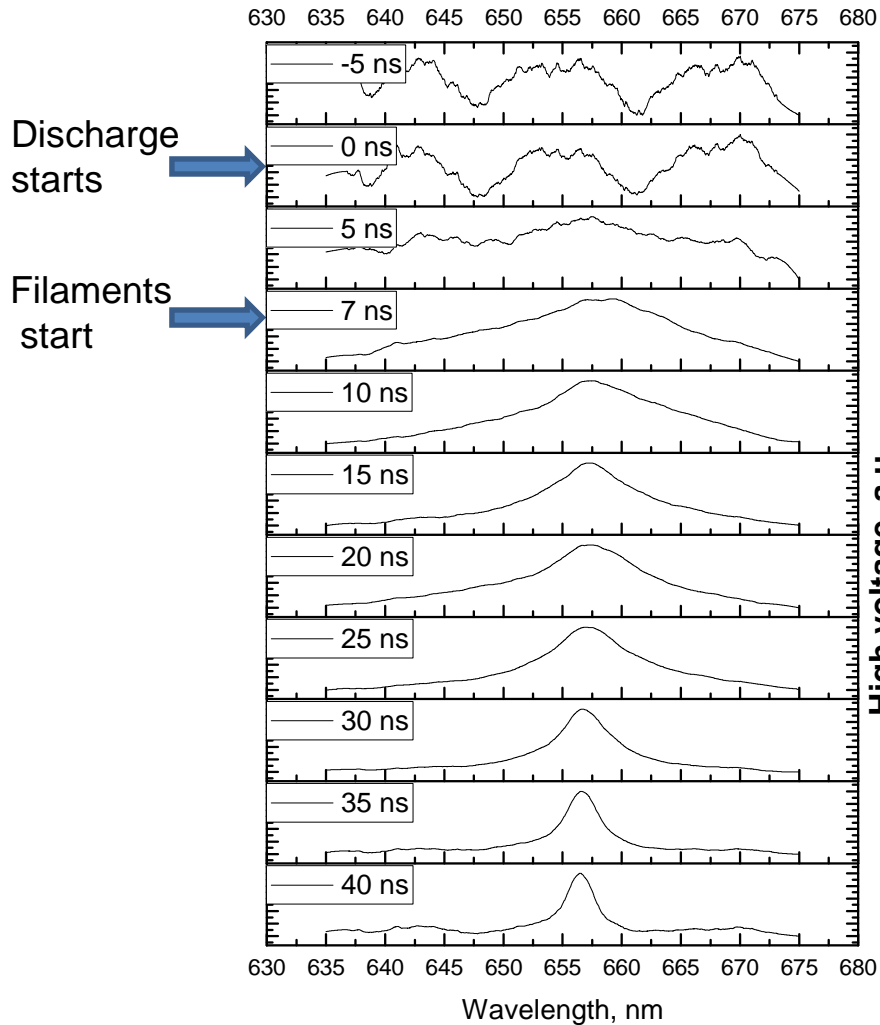
**Single filament is aligned with the entrance slit of the spectrometer**

# Continuous spectra in filamentary discharge

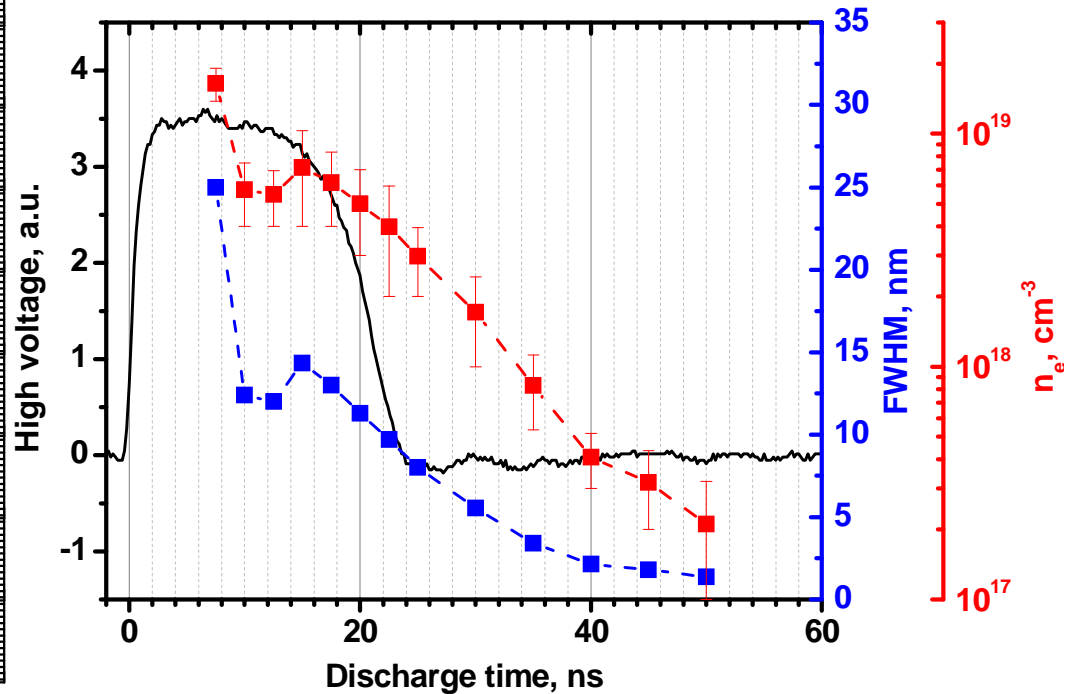




# Broadening of H $\alpha$ line



Filamentary discharge in N $_2$ :H $_2$  (7:1) mixture P=8 bar. Electron density measurements with H $\alpha$  line (656 nm) broadening. Camera gate 5 ns





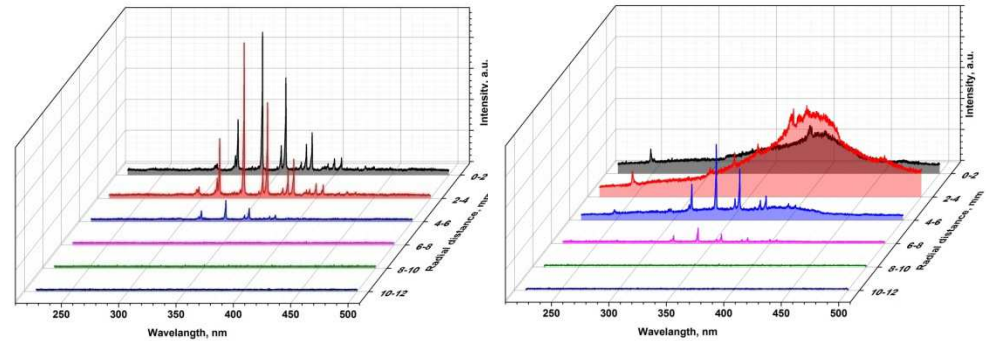
# Recombination emission in the filament



## Electron density from recombination emission

SPS (337.1 nm) is a reference radiation

$$\left\{ \begin{array}{l} N_2(C) = k_C(E/N) \cdot n_e \cdot N_2 / \nu_q \\ Q_C = N_2(C) \cdot F_{FK} \cdot A_{00} \\ Q_C = 3 \cdot 10^{21} \text{ quantum/cm}^3/\text{s} \end{array} \right.$$

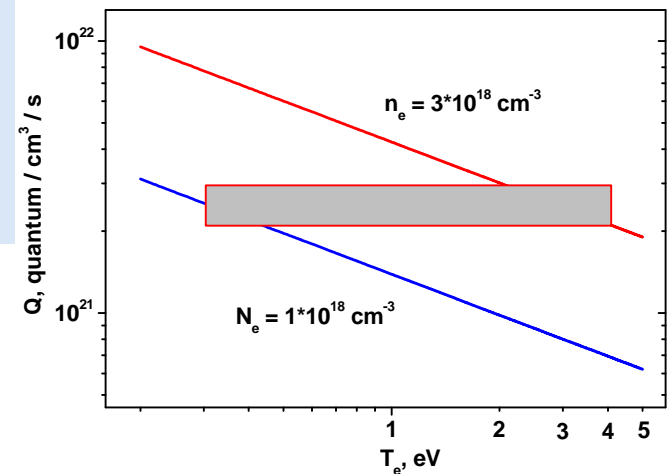


$$Q_C(t=0 \text{ ns}) / Q_{CW}(t=5 \text{ ns}) \sim (1.5-2)$$

$$Q_{CW}(\omega) = C_0 \cdot \frac{n_e \cdot n_{ion}}{\sqrt{T_e} \cdot \eta \omega} d\omega = (1.5-2) \cdot 10^{21} \text{ quantum/cm}^3/\text{s}$$

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

In the discharge  $T_e \sim 2-4 \text{ eV} \rightarrow n_e \sim 3 \cdot 10^{18} \text{ cm}^{-3}$   
 In the afterglow  $T_e \sim 0.5 \text{ eV} \rightarrow n_e \sim 10^{18} \text{ cm}^{-3}$



# Conclusions



Nanosecond dielectric barrier discharge (nSDBD) provides ignition of different fuels in a wide range of stoichiometries

At high gas densities, nSDBD simultaneously provides hundreds of ignition sites distributed in space. The geometry of the ignition and following combustion can be adapted to provide the highest possible efficiency of the system

Physics of filamentation in high pressure nSDBD is under study now ( $n_e=10^{19}$  cm<sup>-3</sup> is achieved during a few nanoseconds in a single-shot mode)