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Calorimetry and Atomic Oxygen LIF of Pulsed Nanosecond Discharges at Above-Atmospheric Pressures

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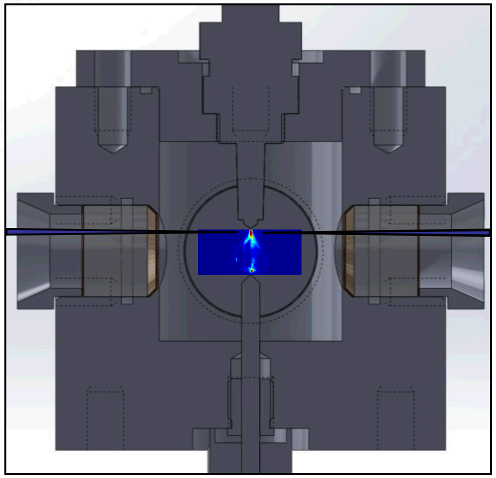

Abstract

The conversion efficiency of secondary electrical energy into thermal energy was measured in air using an optically accessible spark calorimeter for high-voltage (27 kV peak) pulsed nanosecond discharges with secondary streamer breakdown (SSB) and similar low-temperature plasmas (LTP) without. Initial pressures were varied between 1 and 5 bar absolute, with the anode/cathode gap distances like-wise varied between 1 and 5 mm. Secondary electrical energy was measured us-ing an in-line attenuator, with the thermal energy determined from pressure-rise calorimetry measurements. The SSB probability at each initial pressure and gap distance was also recorded. The calorimetry measurements confirm that, similar to inductive spark discharges, SSB discharges promote ignition by increasing the local gas temperature. LTP discharges, on the other hand, had very little local gas heating, with electrical-to-thermal conversion efficiencies of ~1%. Instead, the LTP was found to generate substantial O-atom populations — measured using two-photon laser-induced fluorescence near the anode where electric field strengths were strongest — that persisted for 100's of microseconds after the discharge. The influence of 10 repetitive pulses spaced 100 μs apart was also evaluated for a fixed 5 mm electrode gap distance, with the conditional SSB probability for each pulse evaluated using an available photodiode, with the SSB probability found to have increased for each successive pulse. The influence of chemical and thermal preconditioning by the preceding LTP pulse was evaluated, with the increase in SSB occurrence attributed predominantly to mild gas heating that decreased number densities between the electrodes and hence the gas resistance for the subsequent pulse.

Acknowledgements

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DOE Program Manager: Leo Breton, Gurpreet Singh

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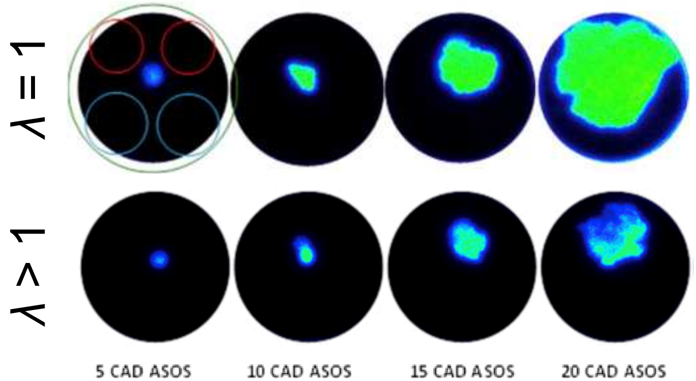
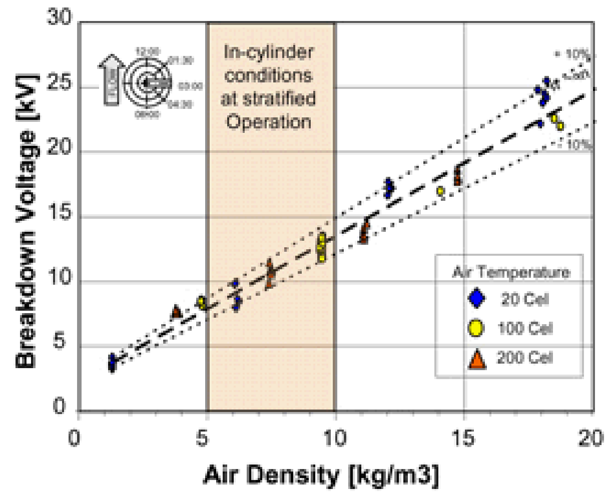
Inductive spark ignition systems have struggled with next-generation gasoline engines due to:

Slow flame kernel growth in dilute charge mixtures



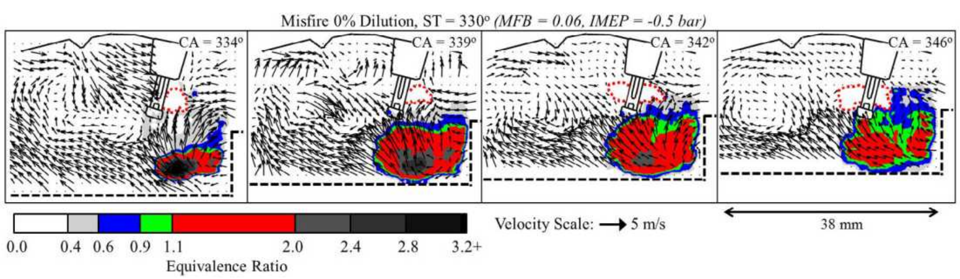
S. Merola, et al, Energy, 2016; 108:50-62.

High breakdown voltages needed for elevated charge densities & large electrode-gap distances



Piock et al, SAE Int J Engines, 2010;3:389-401.

Stratified fuel-air distributions result in intermittently combustible charge mixtures



Peterson, et al, Combust Flame, 2014;161:240-55.

Close-coupled pulsed nanosecond discharges (PND) are a promising ignition strategy for these challenging conditions

Current PND engine research aims for low-temperature plasma ignition, but results confounded by transition to arc

Improved fundamental understanding of next-generation ignition systems, including PND, needed

The goal of this presentation is to

Present (1) calorimetry measurements, including examination of arc transition in PND
(2) O-atom TALIF measurements for single and multi-pulse PND

Background

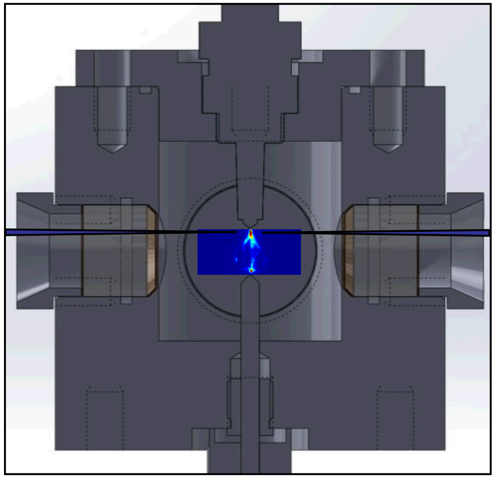
→ Physics of PND
→ Motivation of arc transition problem

Experiment

→ Optically accessible calorimeter
→ Laser/imaging

Results

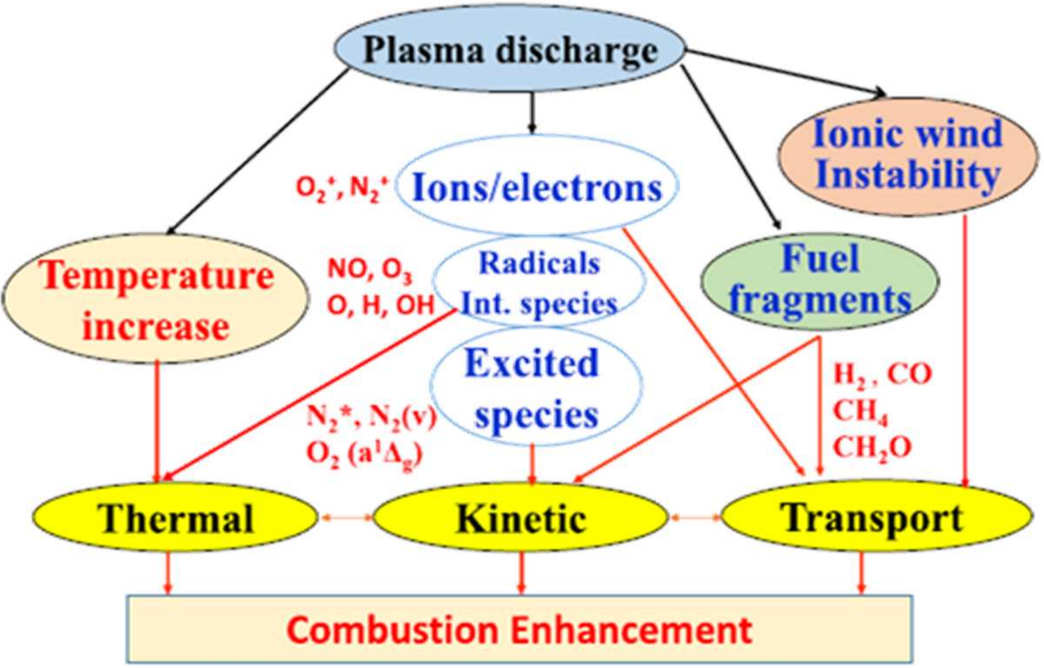
→ Calorimetry/arc transition
Examined gap distance, pressure, number of pulses
→ Factors influencing arc transition
→ O-atom measurements at anode

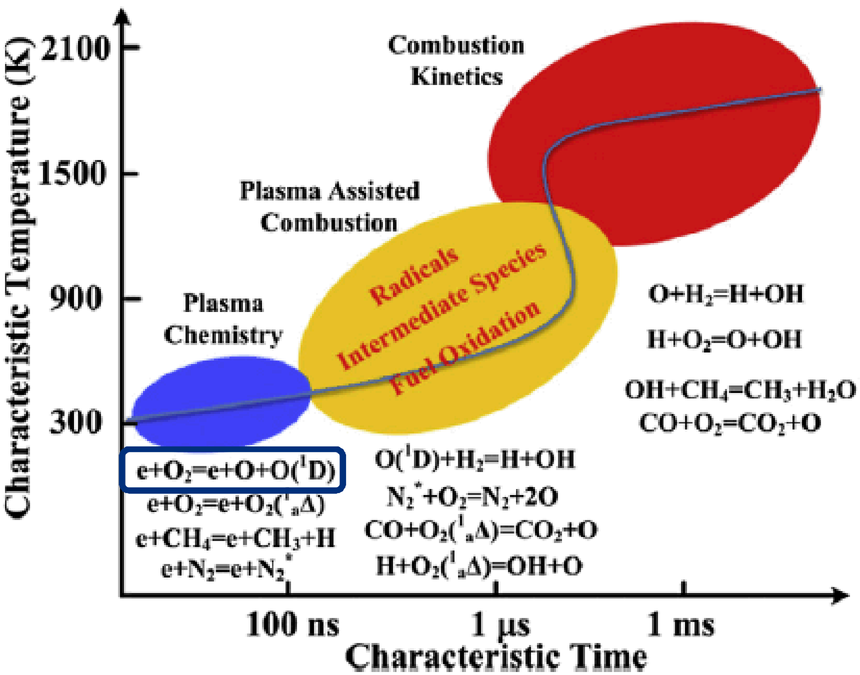


Plasma-assisted combustion (PAC) from pulsed nanosecond discharges (PND) opens new non-thermal chemical pathways

Complex chemical pathways enabled via new phenomenological mechanisms

Shorter plasma timescales relative to ignition kinetics



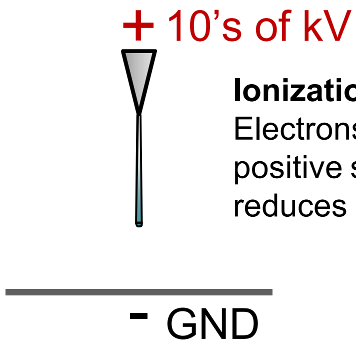


Ju and Sun, *Progr. Energy Combust. Sci.*, 48 (2015) 21-83

Elevated electron temperatures (10s of eV) for PND substantially increase ionization/dissociation rates relative to MW or RF corona ignition

Trick is to have the discharge remain a low-temperature plasma (LTP) so that electrode wear from arc is avoided

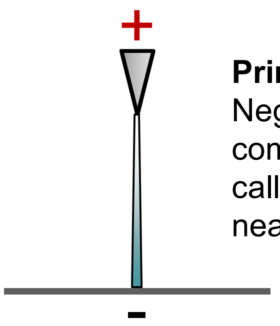
1. Primary streamer phase



Ionization by primary streamer head. Electrons move towards anode leaving positive space charge in streamer path; reduces potential drop near anode.

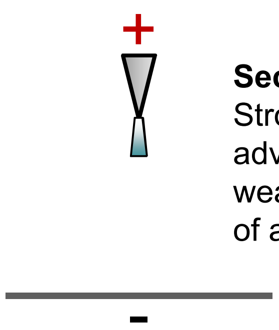
Δt ~ $\mathcal{O}$(1 ns)

2. Compensation phase



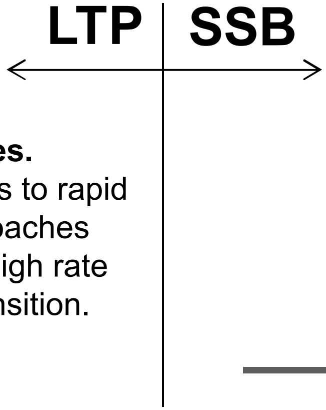
Primary streamer connects to cathode. Negative charge flows from cathode to compensate positive space charge, often called “return wave”. E-field increases near anode, decreases near cathode.

3.Secondary streamer phase

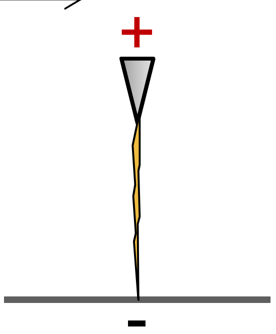


Secondary streamer advances. Strong E-field near anode leads to rapid advance, more slowly as approaches weaker E-field near cathode. High rate of attachment prevents arc transition.

Δt ~ $\mathcal{O}$(10 ns)



4. Transient arc phase



Arc occurs if secondary streamer reaches cathode (*E*/*N* high along the entire discharge filament). Secondary streamer propagation promoted by local heating from V-T relaxation (reduction in *N*).

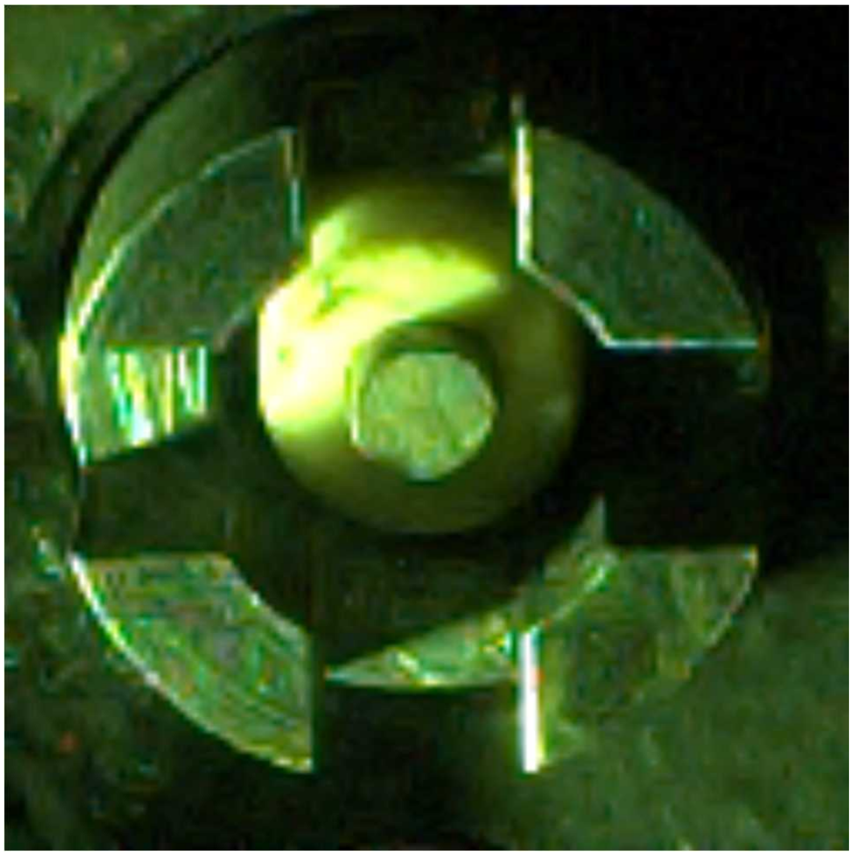
Marode, J. Appl. Phys., 1974;46(5).
Bastien & Marode, J. Phys. D: Appl. Phys., 1985; 18(377).

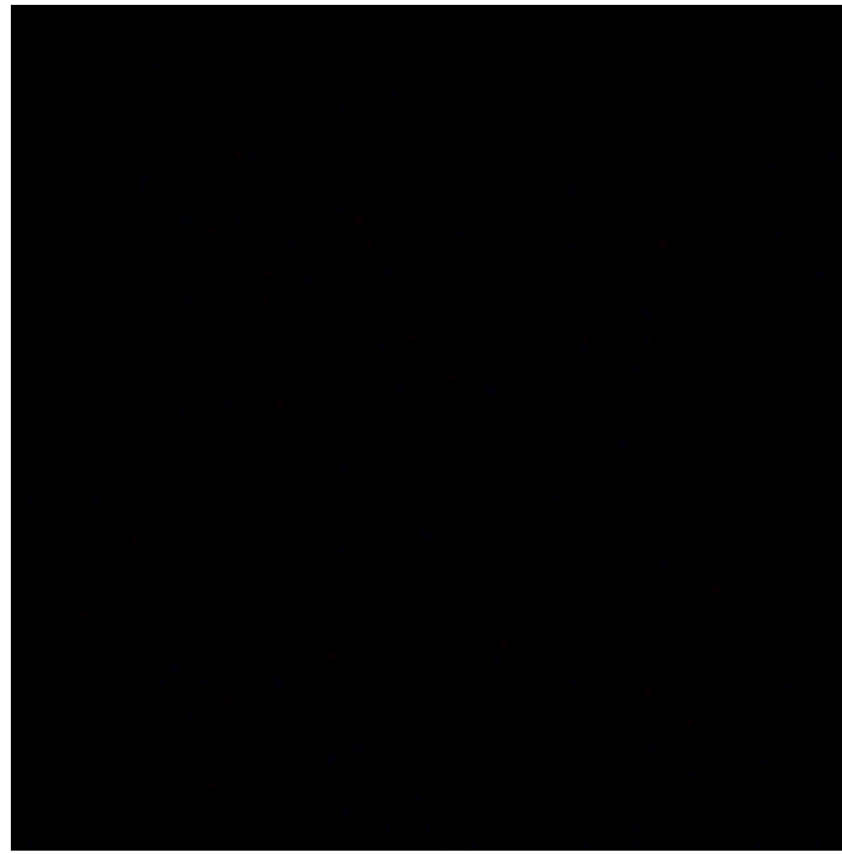
→ Streamer propagation velocity decreases with increased density, reduced applied field

Close-coupled PND (100 μs dwell) often used to increase radical formation while ideally avoiding secondary streamer breakdown (SSB)

Limited optical evidence suggests that multi-pulse PND leads to SSB for conditions where single-pulse PND does not

Sjöberg et al. *SAE Int. J. Engines*, 7 (4):1781-801, 2014

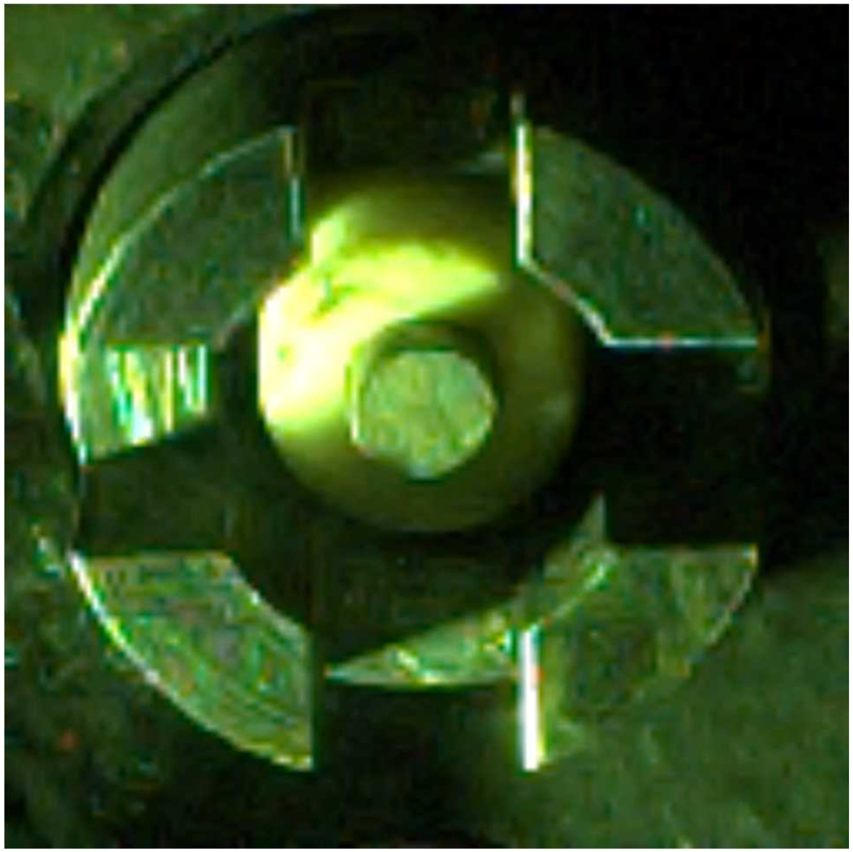




- Once SSB occurs in the pulse-burst, it occurs for each subsequent pulse
- Successive SSB events are biased to the same cathode site

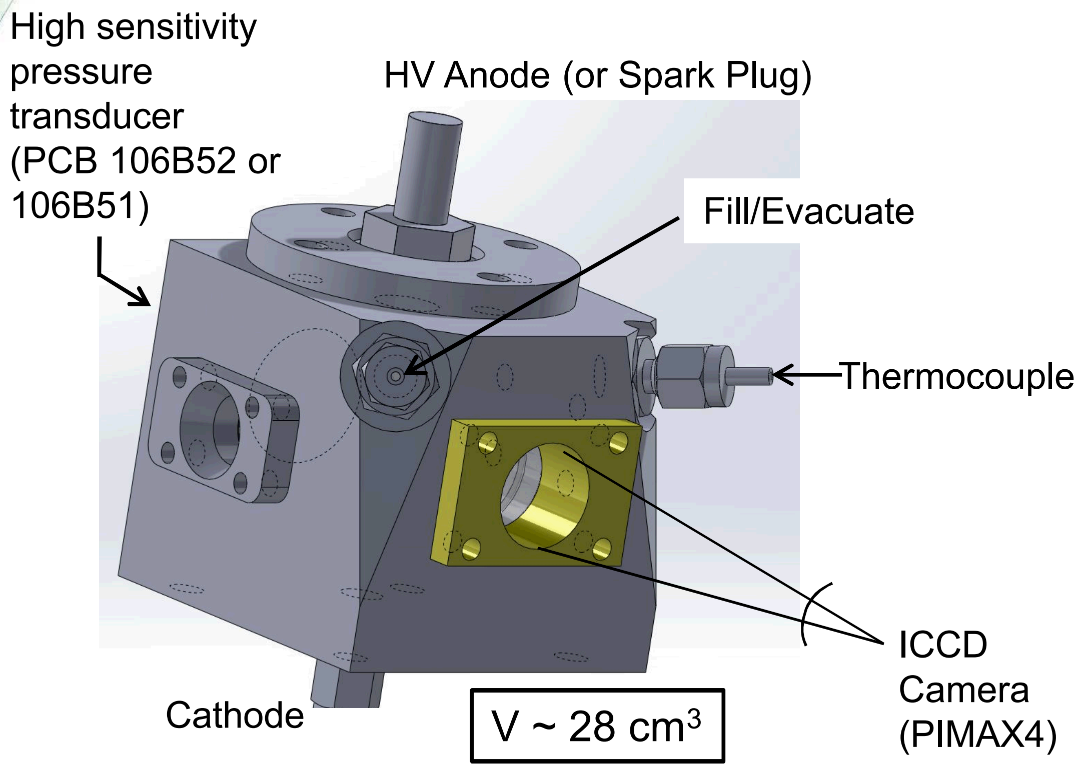
Limited optical evidence suggests that multi-pulse PND leads to SSB for conditions where single-pulse PND does not

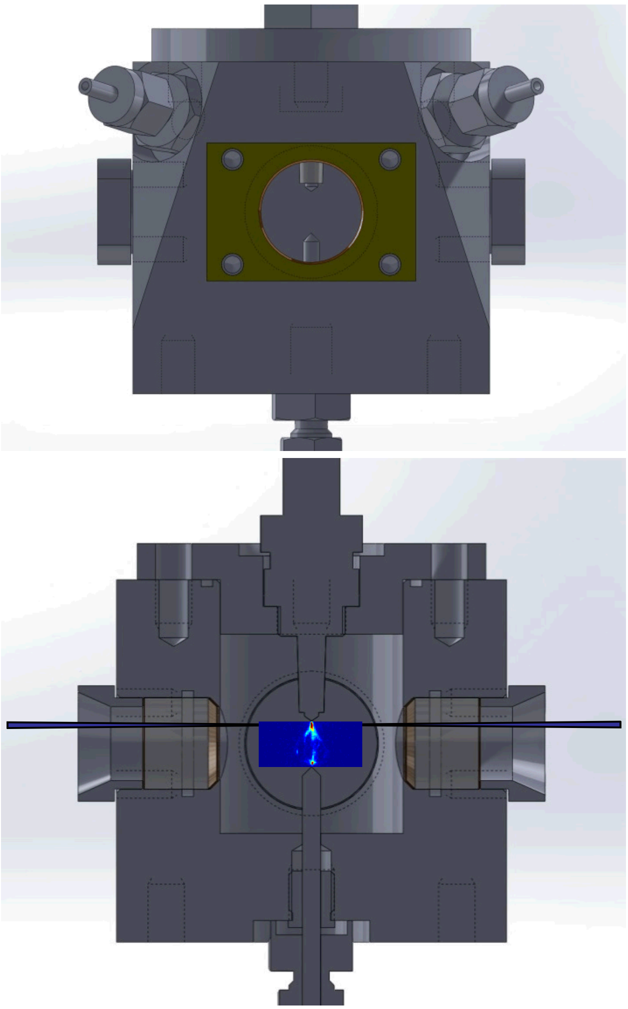
Sjöberg et al. *SAE Int. J. Engines*, 7 (4):1781-801, 2014



Observations suggest some form of thermal or chemical inter-electrode gas preconditioning by the preceding pulses

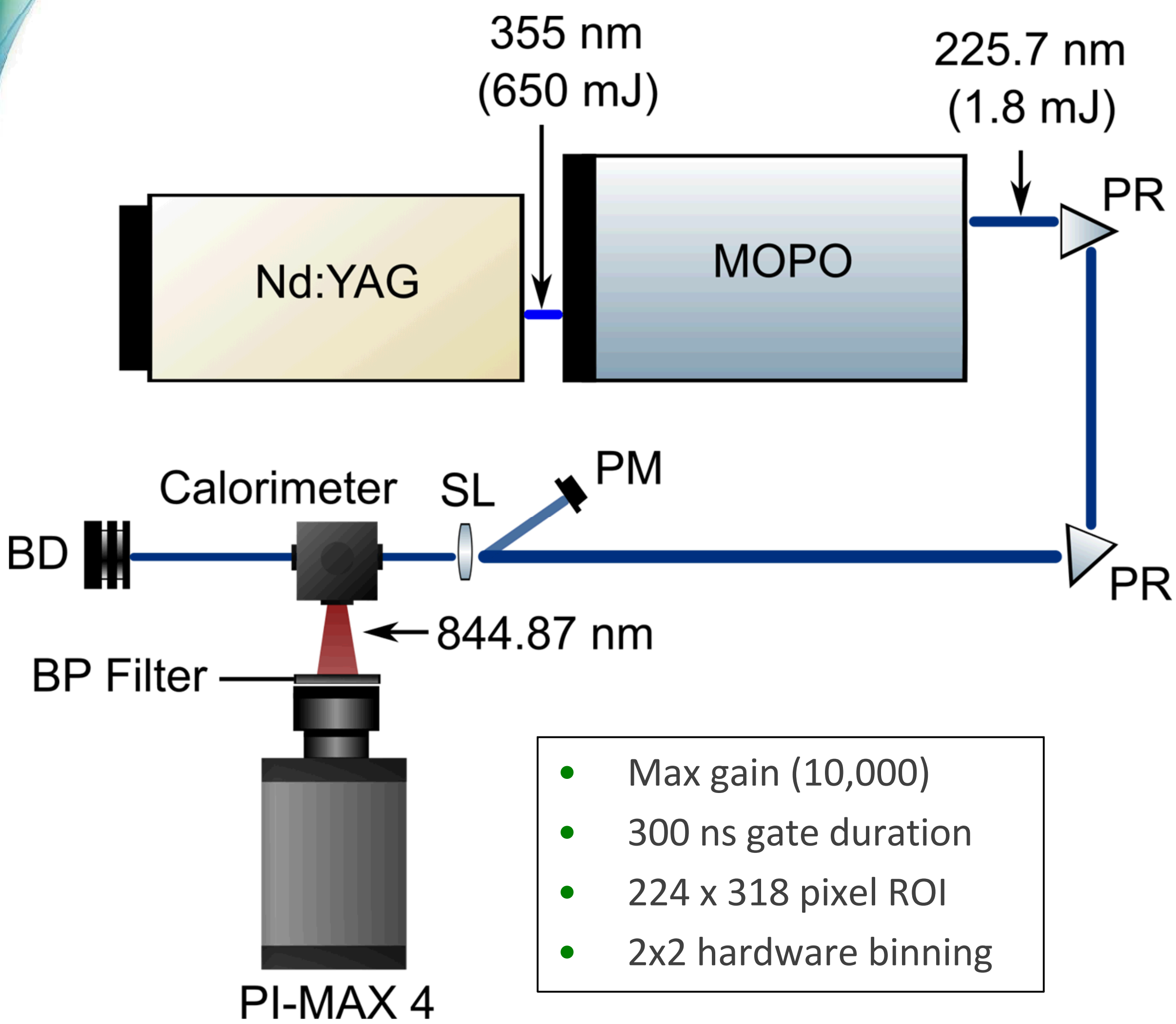
Optically accessible chamber for simultaneous O-TALIF and pressure-rise calorimetry





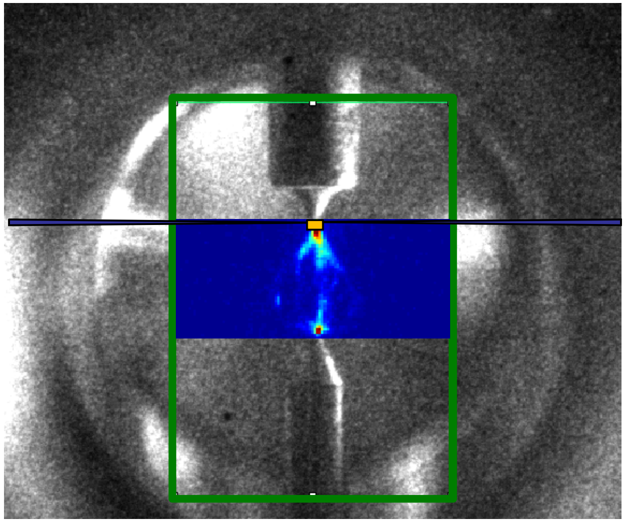
- PCB 106B52 transducer limited to 4.4 bar (725 µV/Pa)
- Incorporated 106B51 transducer with 34 bar limit (145 µV/Pa)
- Pressure control valve (Proportion-Air) for accurate chamber fill pressure control up to 10 bar

Nd:YAG pumped MOPO provides 225.7 nm beam at 1.8 mJ/pulse for O-TALIF



- PR UV Prism
- PM Shot-resolved power meter
- SL 100 mm FL, plano-convex spherical lens
- BD Beam dump
- BP 850 ± 40 nm filter (99.7% @ 845 nm)

- Max gain (10,000)
- 300 ns gate duration
- 224 x 318 pixel ROI
- 2x2 hardware binning



Experiments conducted in "ultra-zero" air, O-TALIF performed at range of delays after discharge

- In-house pulse generator

Transient Plasma Systems

Voltage setting dial = 10 incr. (V_{peak} ~ 27 kV)

Number of pulses = 1, 2, 5, 10

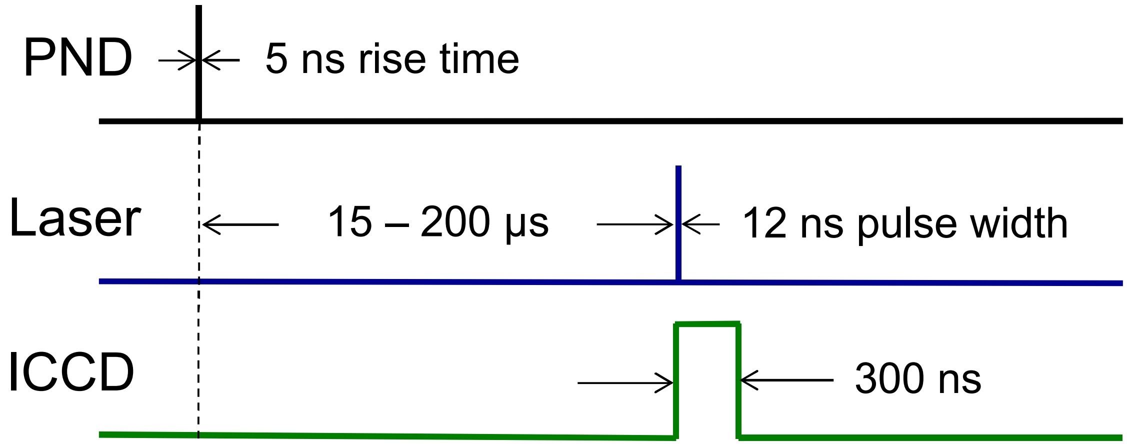
Pulse Δt = 100 μs, 200 μs, 500 μs, 1 ms

Air, Ultra Zero Certified

Guaranteed Specifications

Carbon Dioxide	<	1 ppm
Carbon Monoxide	<	1 ppm
THC	<	0.1 ppm
Water	<	2 ppm
Oxygen		20-22 %

Purge and fill between each run ($1\frac{1}{2}$ min. w/ pressure transducer)



Modified NGK DP7EA-9 spark plug, non-resistor, 1 – 5 mm pin-to-pin gaps

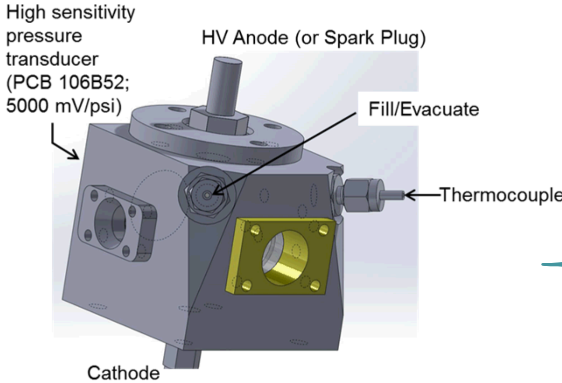
Parametric pressure-rise calorimetry study of PND at range of initial pressure and electrode gap distances

Electrode gap distance
1.0 – 5.0 mm, pin-to-pin

Initial pressure
1.07 – 5.0 bar

TPS pulser setting
Maximum (27 kV_{peak})

Number of pulses
1, 2, 5, & 10 (Δt = 100 μs)



~30 runs
at each condition

Probability of SSB vs. LTP

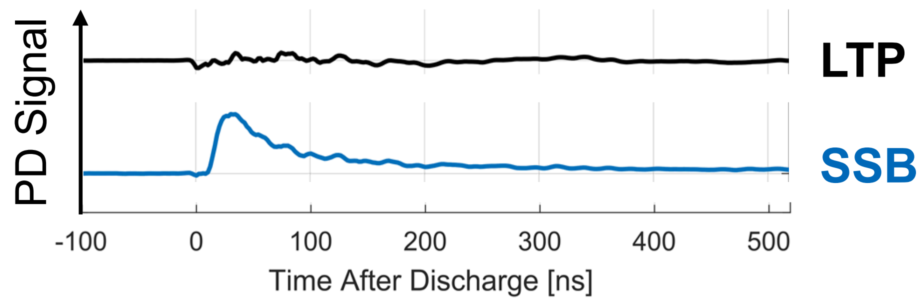
Electrical-to-thermal efficiency, conditional

Electrical energy delivered to electrode gap, conditional

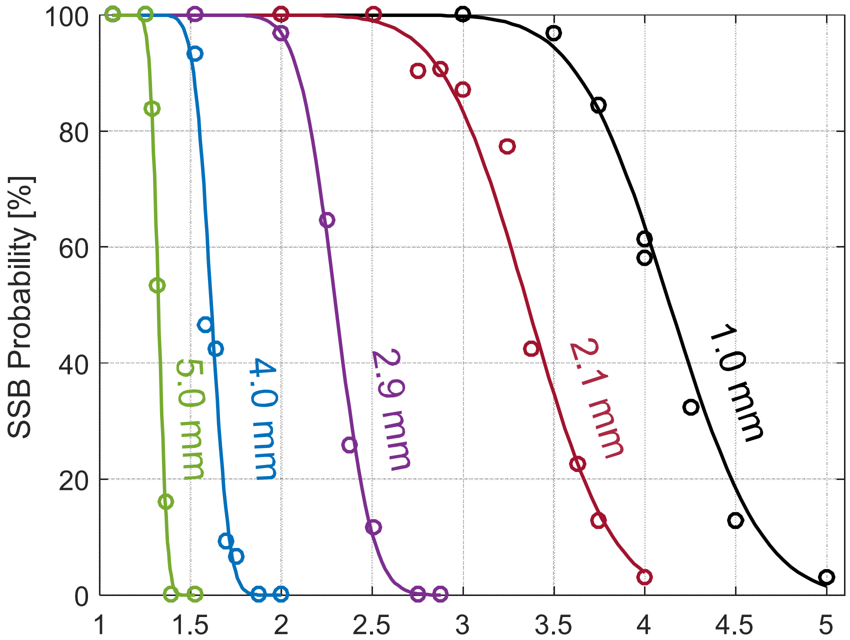
Single- vs. multi-pulse SSB probability, conditional

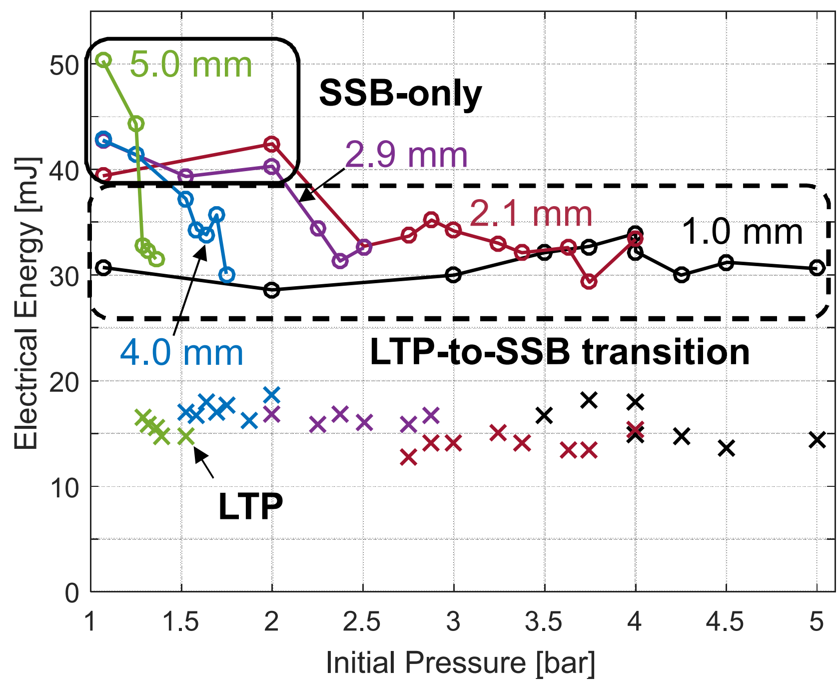
High-speed photodiode monitors light emission from discharge

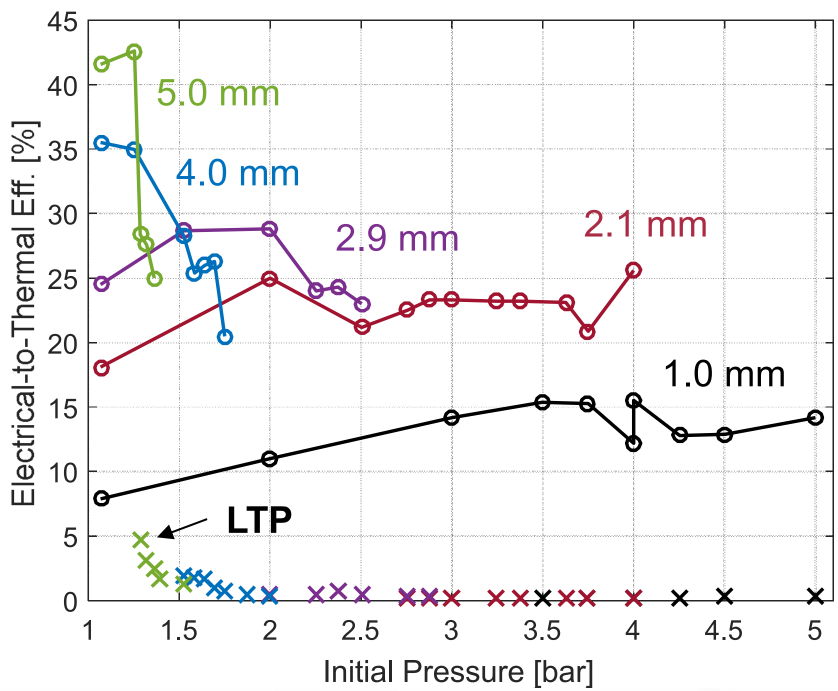
High-speed Photodiode (Thorlabs DET210)



PD signal can be compared to voltage and current profiles to determine when transition to SSB occurs







Single-pulse PND results

- Increasing pressure or gap distance
  - Decreases SSB probability
  - Increases SSB efficiency

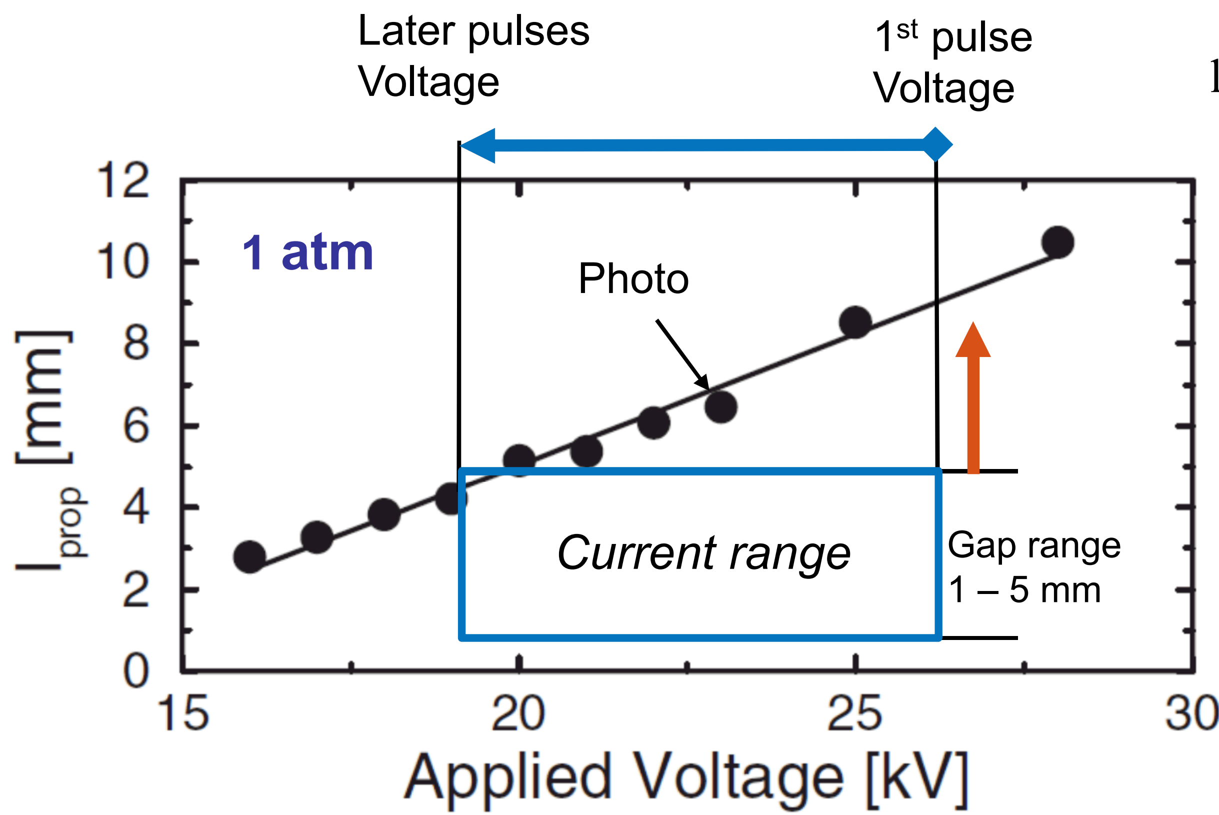
Expected

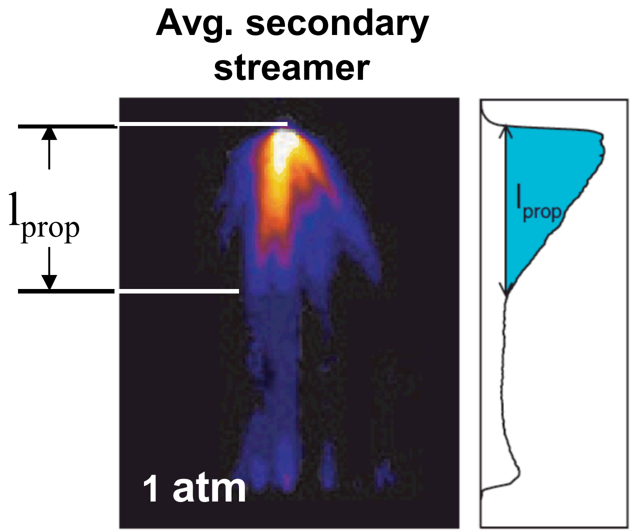
- LTP-to-SSB transition smoother for smaller gap distances
  - Likely due to more consistent electrical energy (physical reason not known)

Unexpected

- SSB efficiency increases with gap size
  - LTP efficiency generally below 1%
  - Small sample size leads to some noise

Secondary streamer likely propagates across gap in our experiment at 1 bar; consistent with observation of SSB



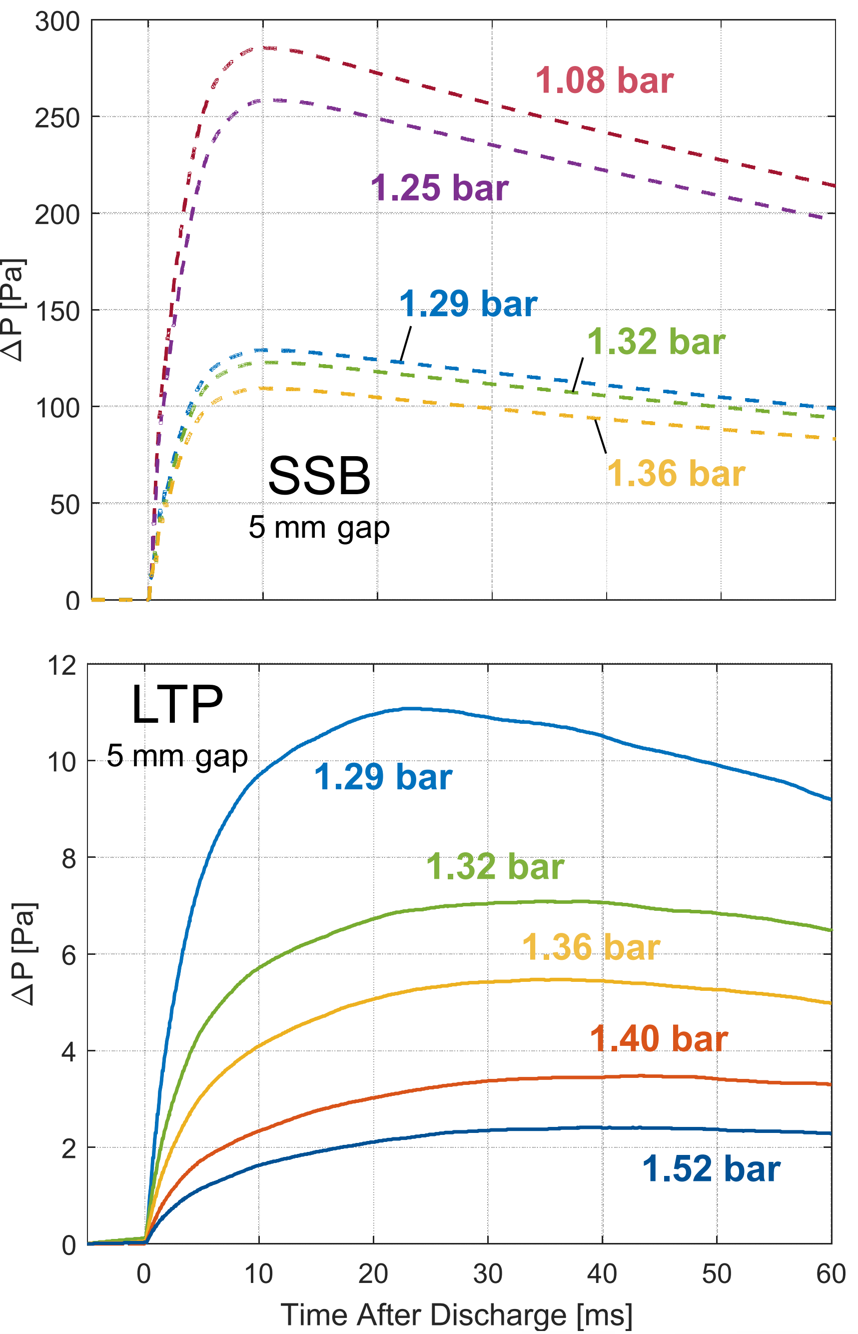


$l_{prop} > d_{gap}$

Suggests secondary streamers cross gap in our experiments at atmospheric pressure

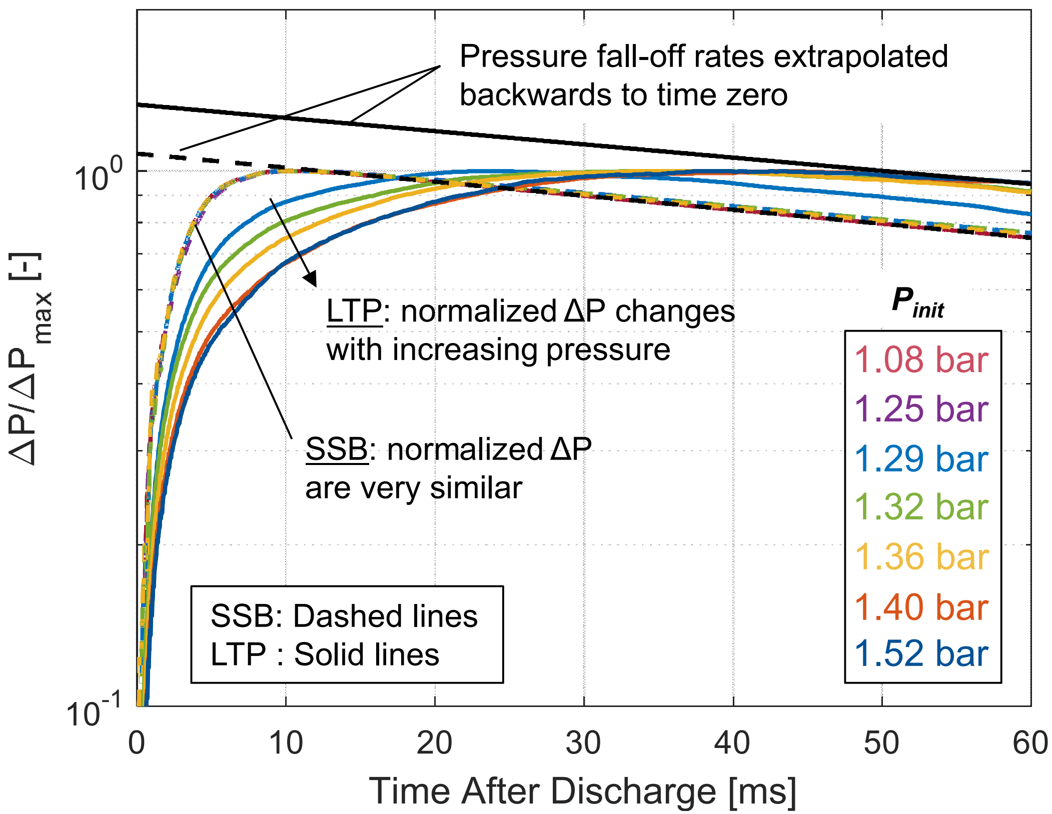
Ono, et al, J. Phys. D: Appl. Phys., 2003;36:1952-58.

Pressure fall-off rates can be used to correct for heat losses; 8% for SSB, ~30% for LTP

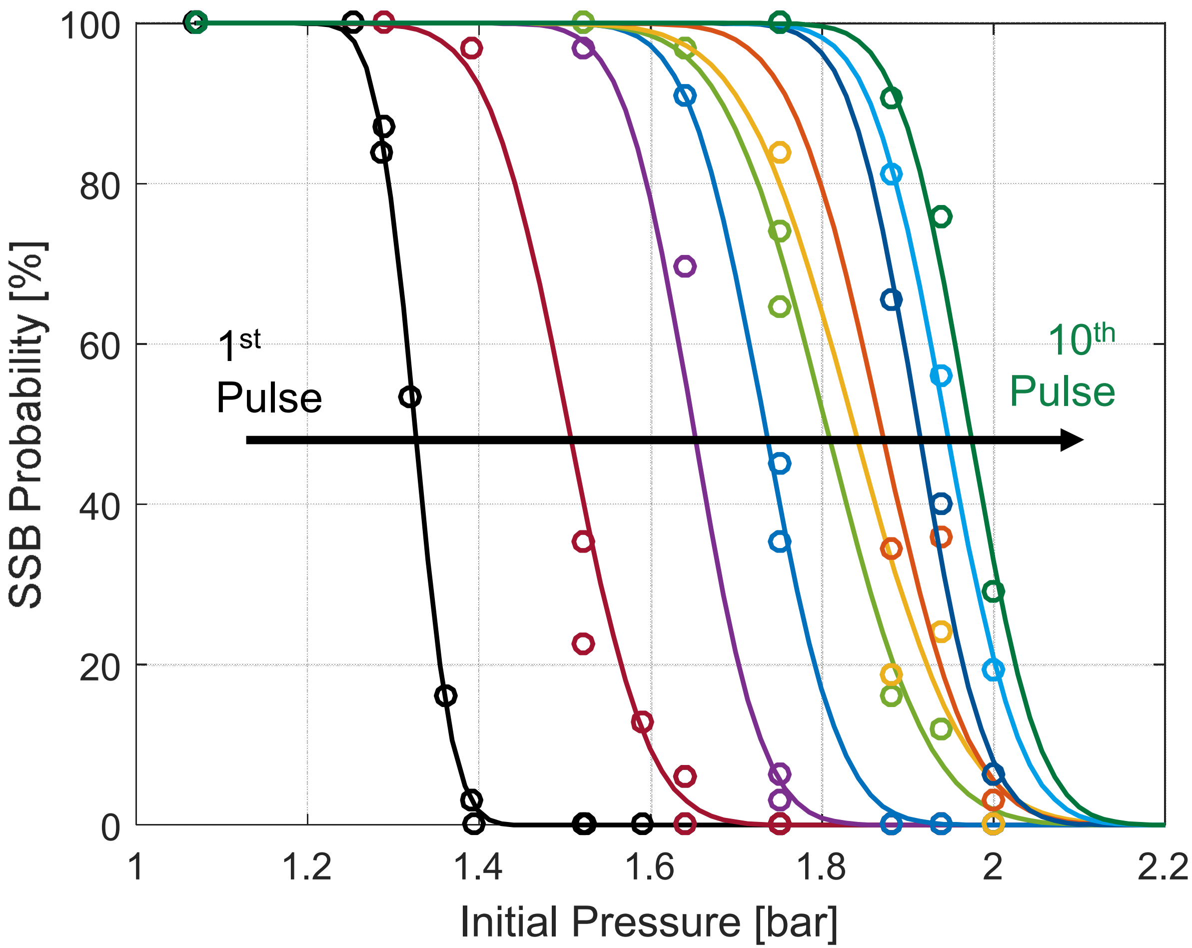


→ Post discharge normalized pressure fall off (heat loss) consistent among both discharge types

Slope { SSB = 2.62e-3; LTP = 2.95e-3 }



Increasing number of pulses extends SSB probability to higher pressure, supports "pre-conditioning" of gas in gap



What physical mechanism "pre-conditions" the gap?

Chemical

- Lower ionization potential ground-state species
- Electronic excited states

Thermal

- Local heating reduces number density (increases E/N)

Gap = 5.0 mm

Evaluation of the potential chemical effect on gas pre-conditioning

Two possible sources

Higher electronic excited state species

- Most excited states w/ half-life $\mathcal{O}$(~ns)
- Singlet oxygen – O_2(a1)
  - meta-stable w/ characteristic half-life of 1e4 seconds in air at 1 bar, 300 K
  - O_2 IP drops 1 eV (~8%)

Lower ionization potential (IP) species post-discharge (e.g. NO)

- N-atom IP ~6% lower than N_2 value
  - O-atom IP ~12% higher
  - Number density doubles (lower E/N)
- NO IP much lower than N_2/O_2 values

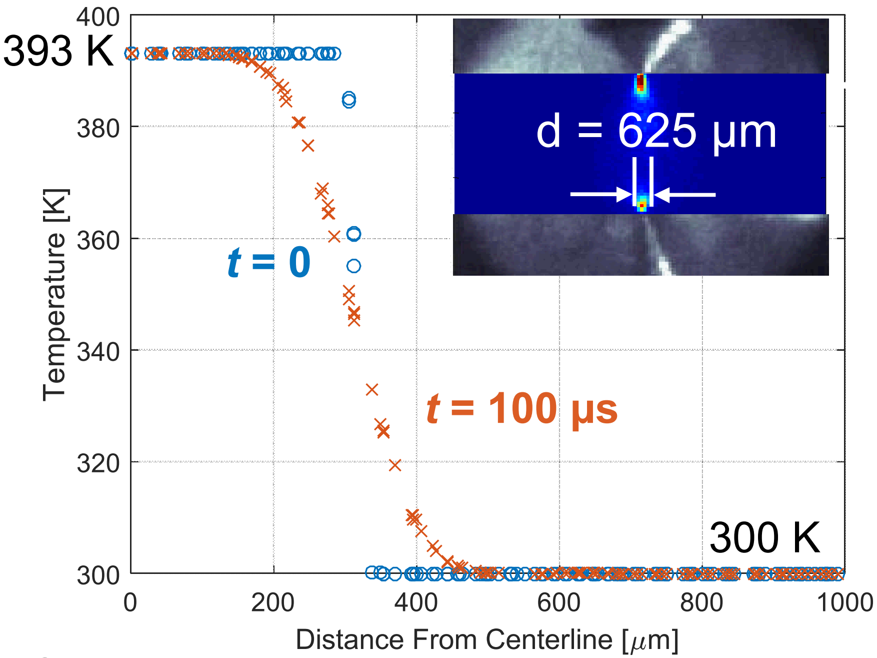
Species	IP [eV]
O_2	12.1
N_2	15.6
NO	9.3
O_3	12.5
O	13.6
N	14.5
O_2(a1)	11.1

- NO or O_2(a1) seem to be the only LTP generated species that could impact subsequent discharges
  - Minor impact from other species
  - Discharge in pure N_2 would remove these effects
  - Competing effects from increased number density
- Suggests chemical effect is important but not dominant

Evaluation of the potential thermal effect on gas pre-conditioning

Temperature increase from LTP

Thermal diffusion 100 μs later
Matlab PDE Toolbox:
1D Heat Equation (Cylindrical)



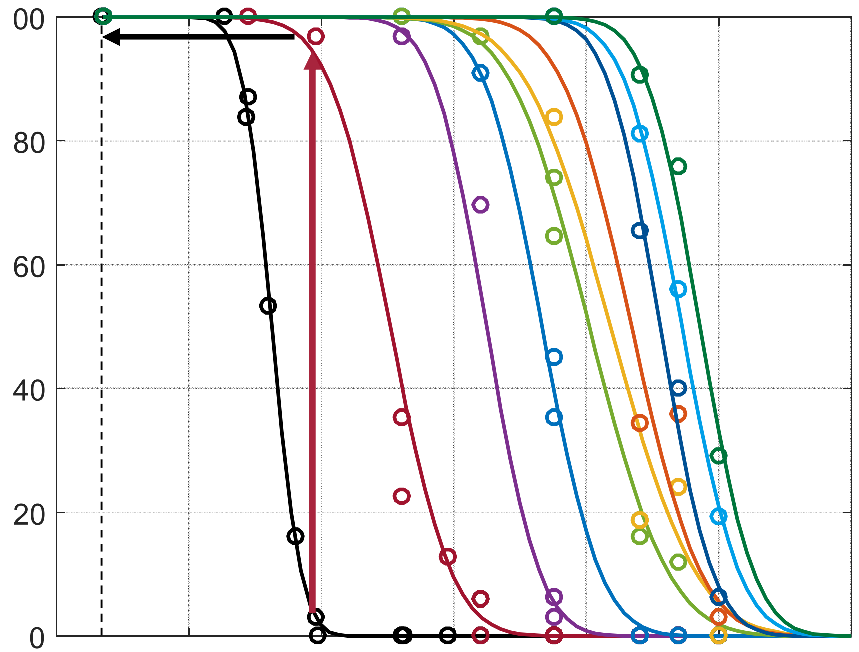
Gap = 5.0 mm
P_{init} = 1.4 bar
$E_{thermal}$ = 240 μJ

$$\Delta T = \frac{E_{Thermal}}{\rho c_p V}$$

→ From calorimetry

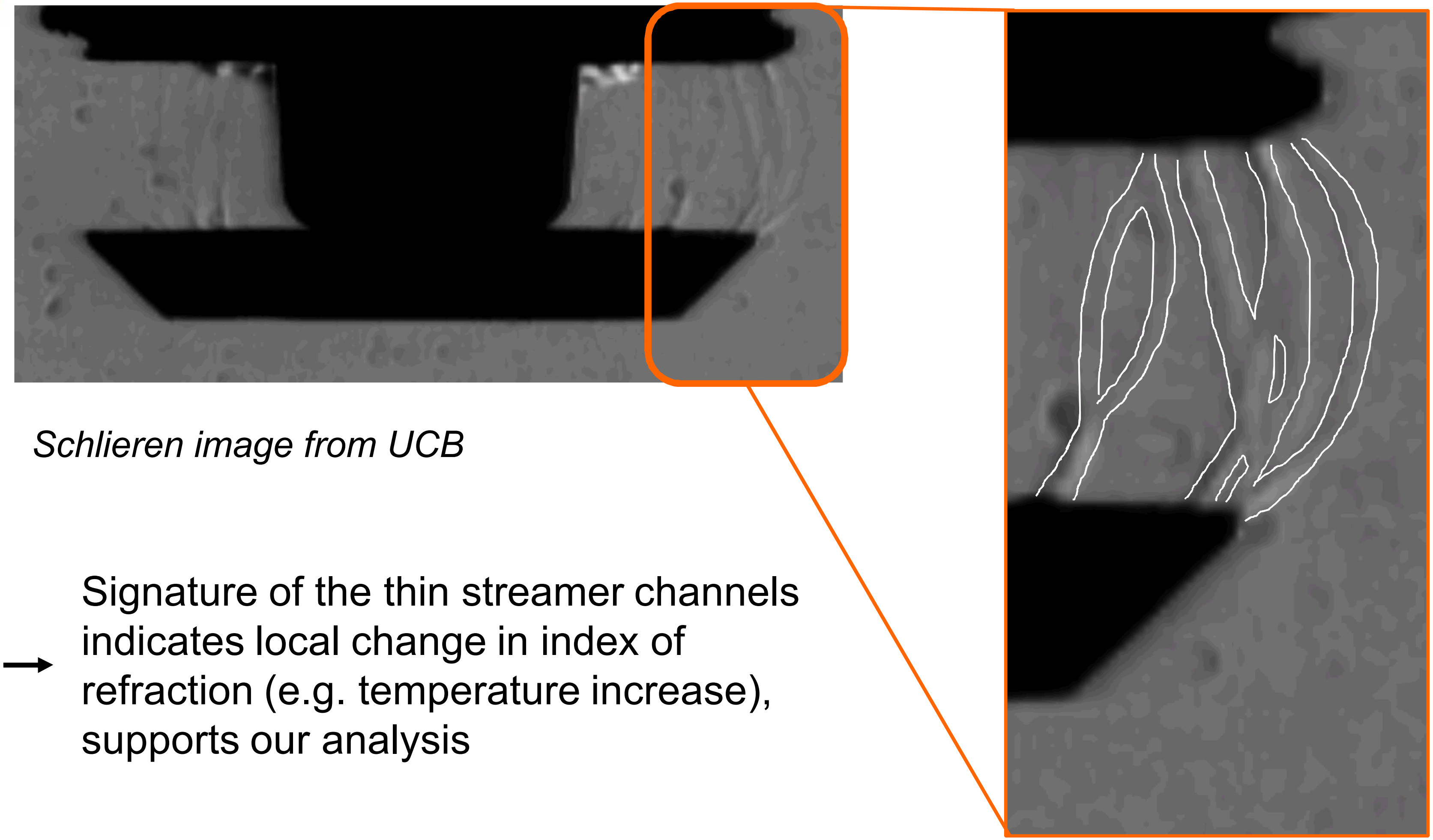
→ Assume cylinder between electrodes, radius from direct imaging/O-PLIF

- 30% increase in centerline temp.
  - 2^{nd} pulse has equivalent decrease in number density: 1.4 bar → 1.07 bar



Thermal effect appears to be dominant mechanism

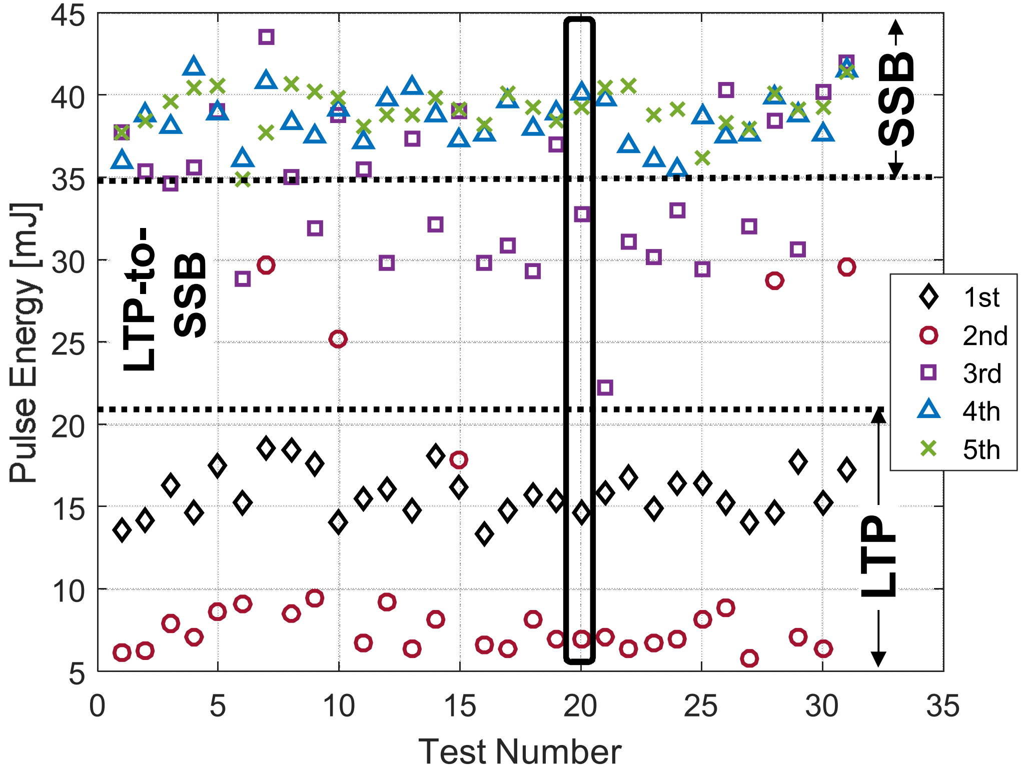
Schlieren imaging performed at UCB shows "hot" streamer channels in LTP discharge

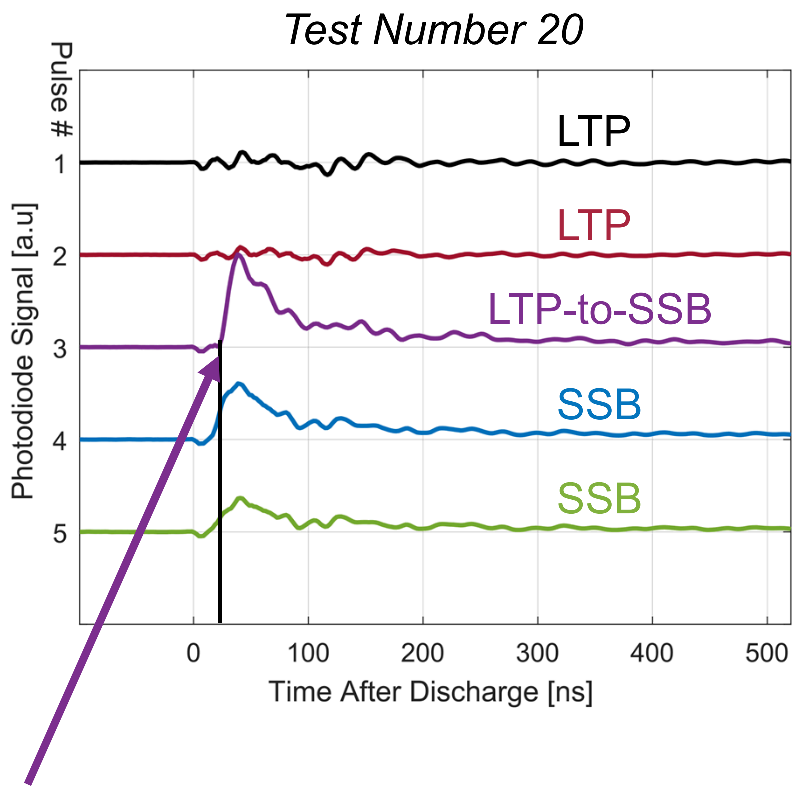


Schlieren image from UCB

→ Signature of the thin streamer channels indicates local change in index of refraction (e.g. temperature increase), supports our analysis

Sequence datasets provide some insights into discharge regimes: LTP, SSB, and transition LTP-to-SSB



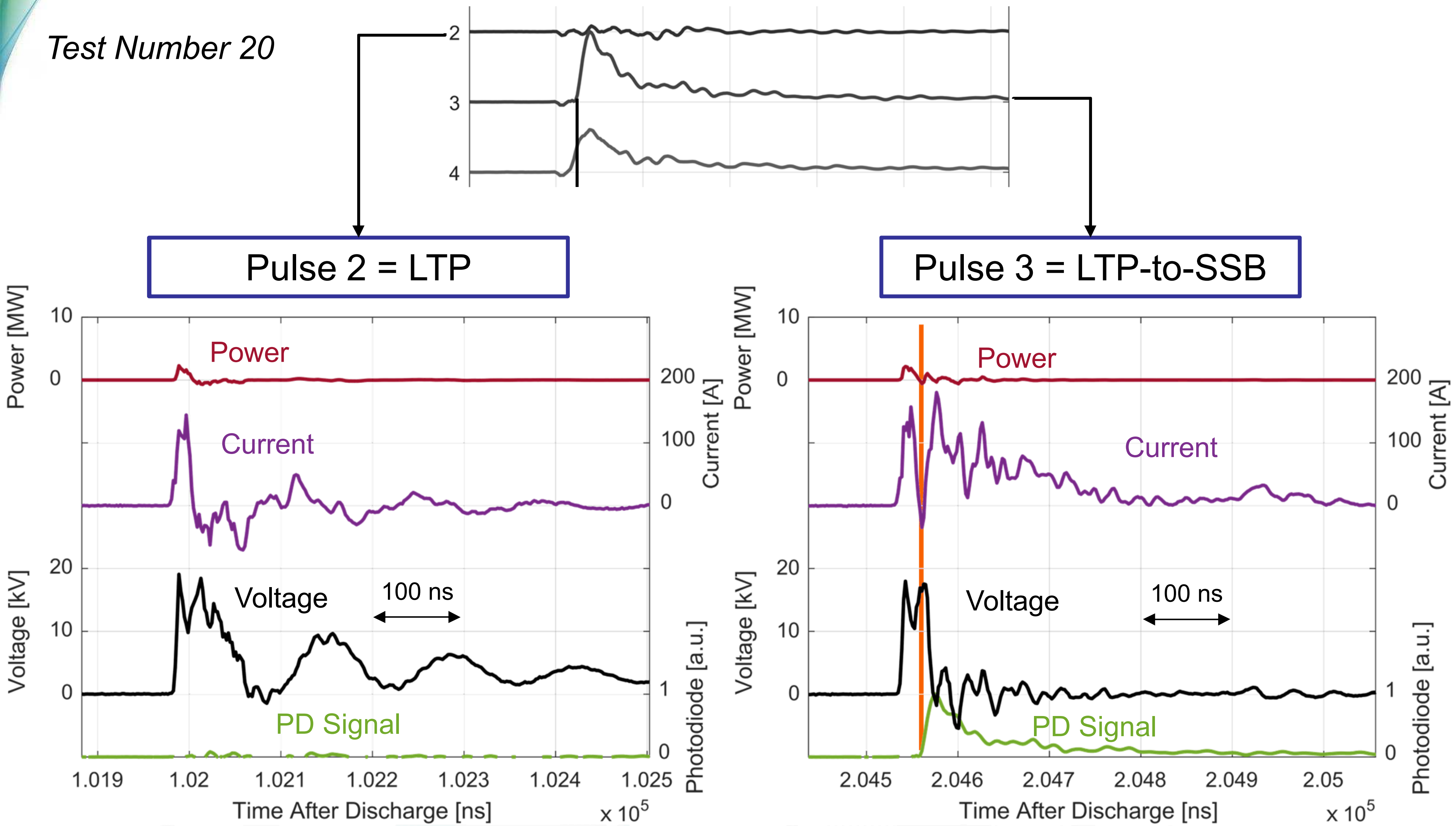


SSB occurrence delayed by ~15 ns

Gap = 5.0 mm
P_{init} = 1.64 bar

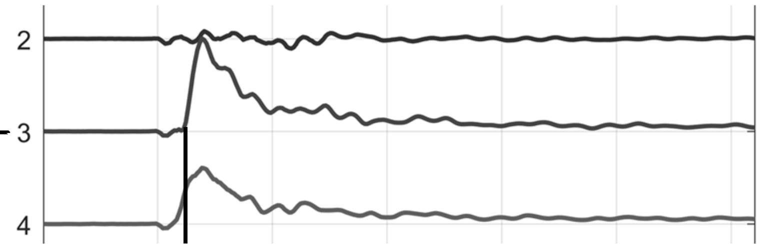
SSB depends on secondary streamer velocity, can occur after current drops

Test Number 20



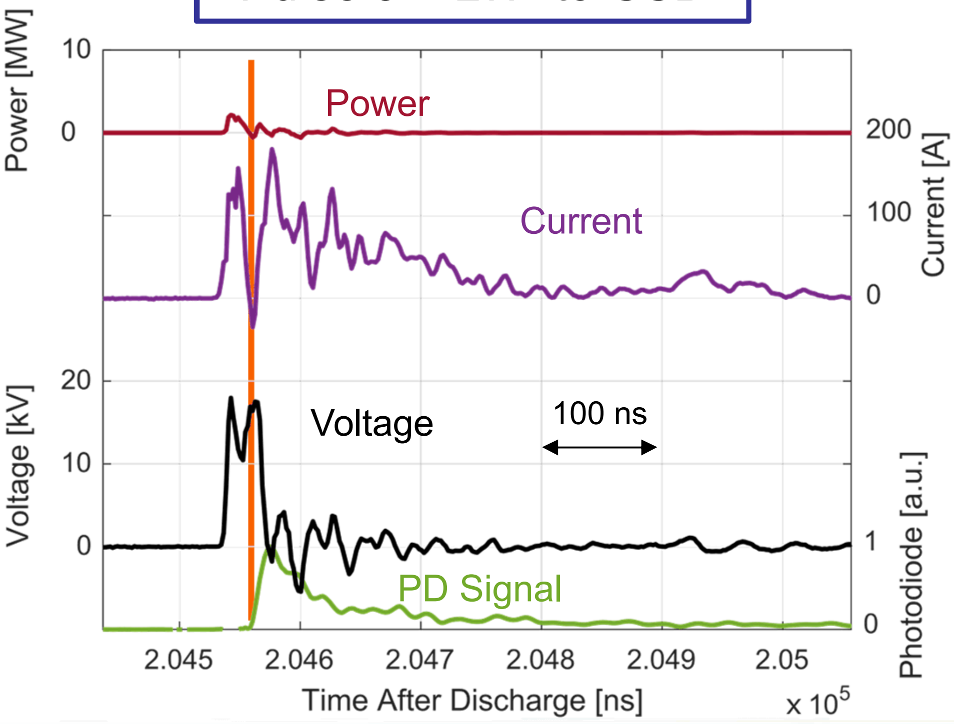
Faster streamer propagation: SSB can occurs near current peak

Test Number 20

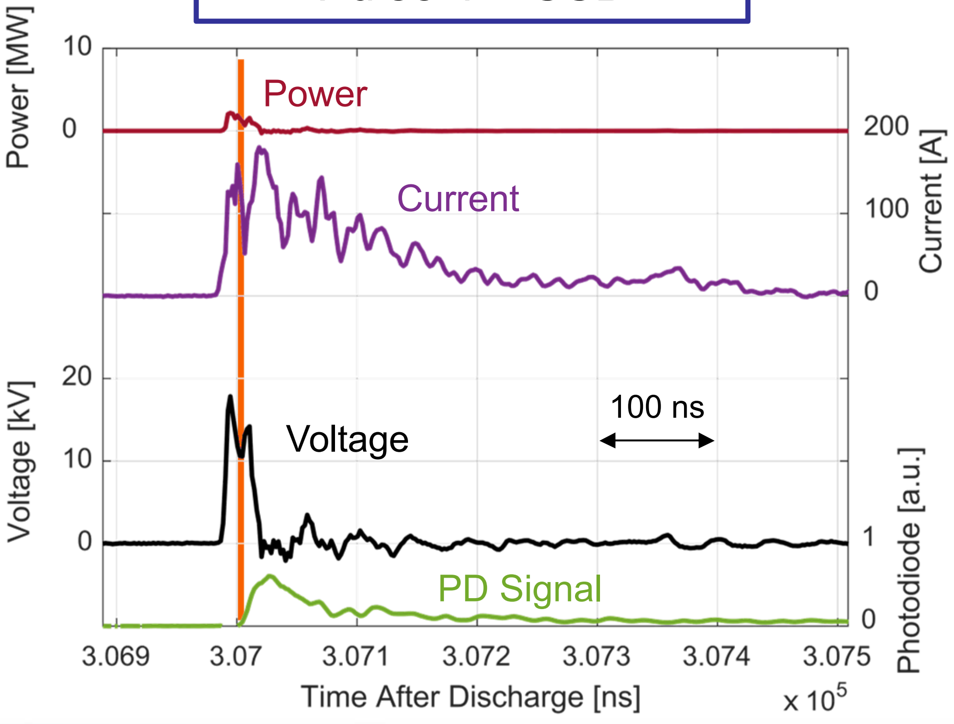


Secondary streamer propagation slower for Pulse 3 compared to Pulse 4

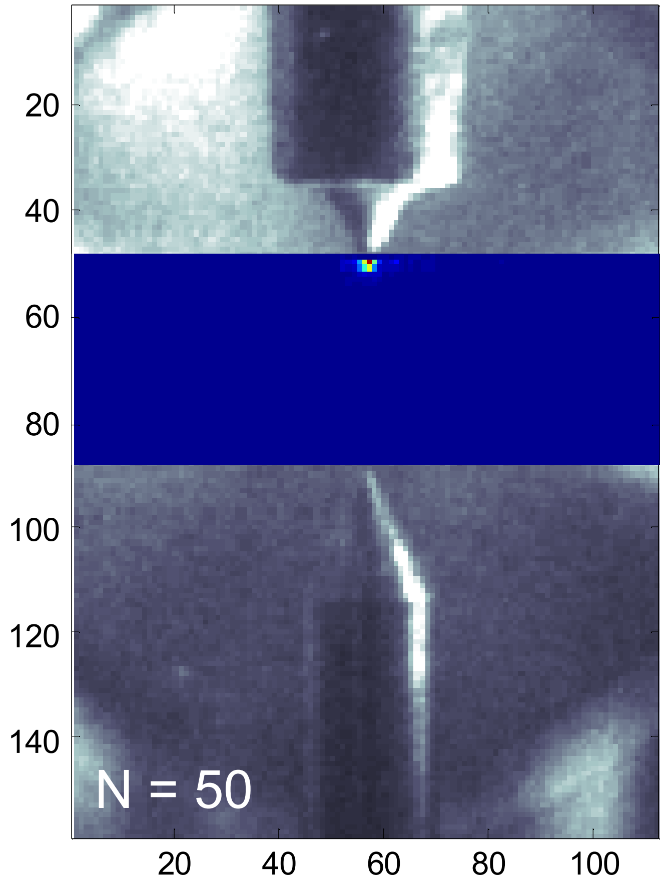
Pulse 3 = LTP-to-SSB

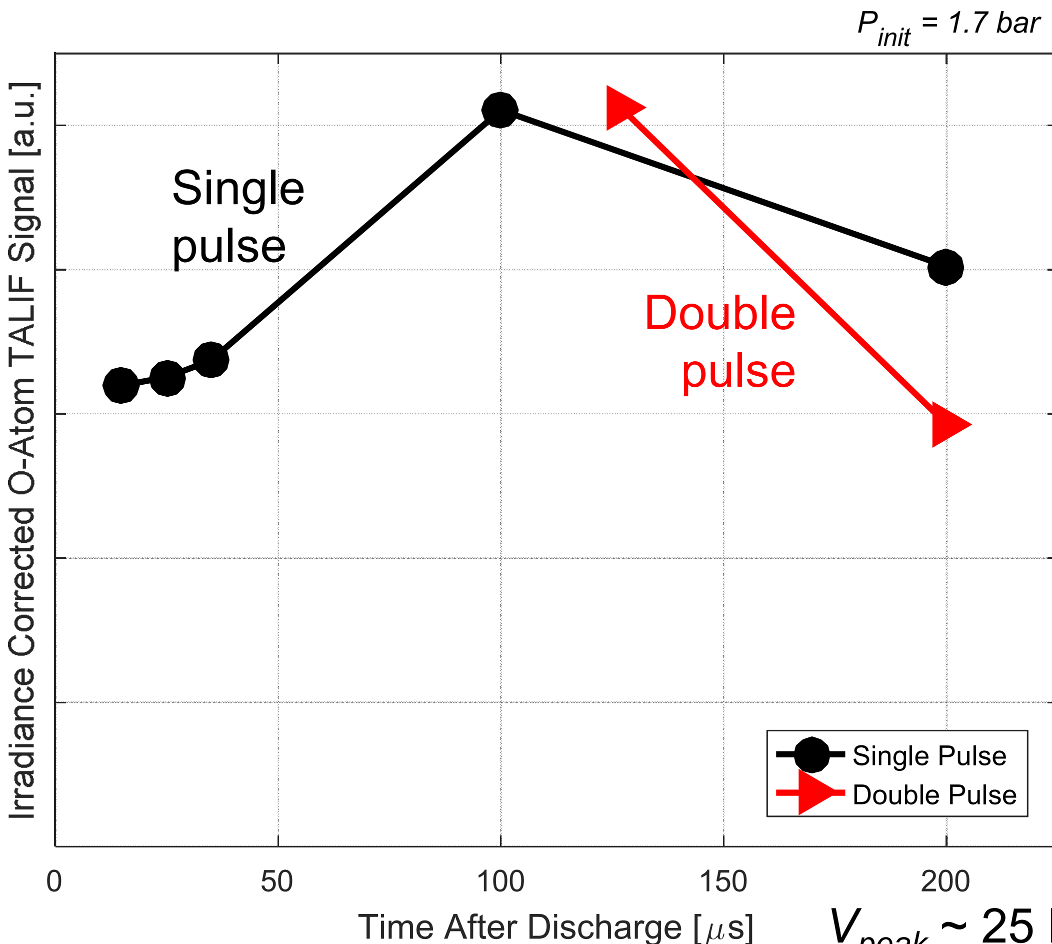


Pulse 4 = SSB



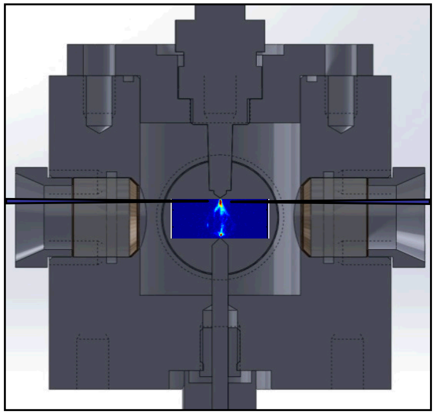
O-atom TALIF signal increases for ~100 µs after discharge, second pulse has negligible impact





V_{peak} ~ 25 kV
Gap = 5 mm

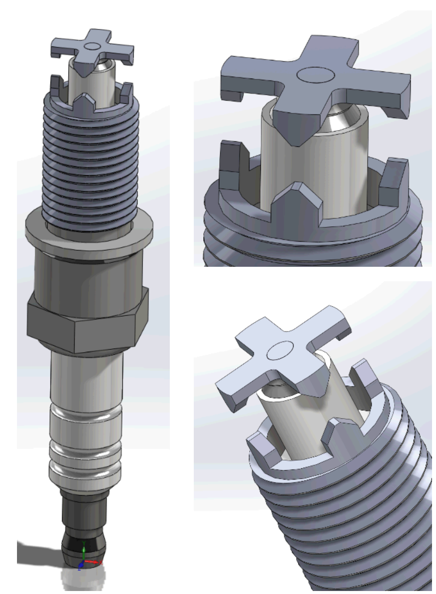
Local heating along streamer channels identified as likely cause of arc transition



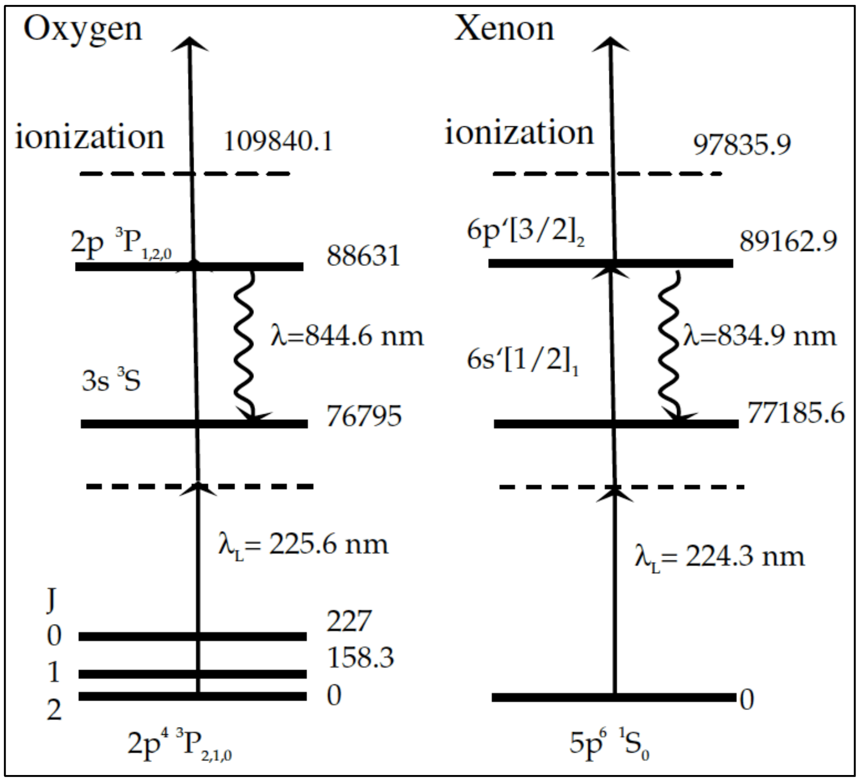
- Decreased pressure and gap make SSB more likely
- For multi-pulse, “slight” temperature increase from early pulses decreases number density sufficiently for SSB to occur
- Increased gas refresh rate could reduce SSB likelihood on successive pulses: open electrodes, increased gas velocity
- O-TALIF measurements indicate increasing O-atom concentration for 100 µs after discharge; second pulse has minimal impact

Next steps…

- Increased pressure and temperature, capable of up to 9 bar
- Effects of various molecular constituents: CO2, water, hydrocarbons
- Direct imaging of O and N2 relaxation to ground state
  - Electronic excitation of ground state O and N2 by electron impact
- Test custom spark plug that can be installed in optical engine



In-situ calibration with xenon leads to absolute O-atom number density



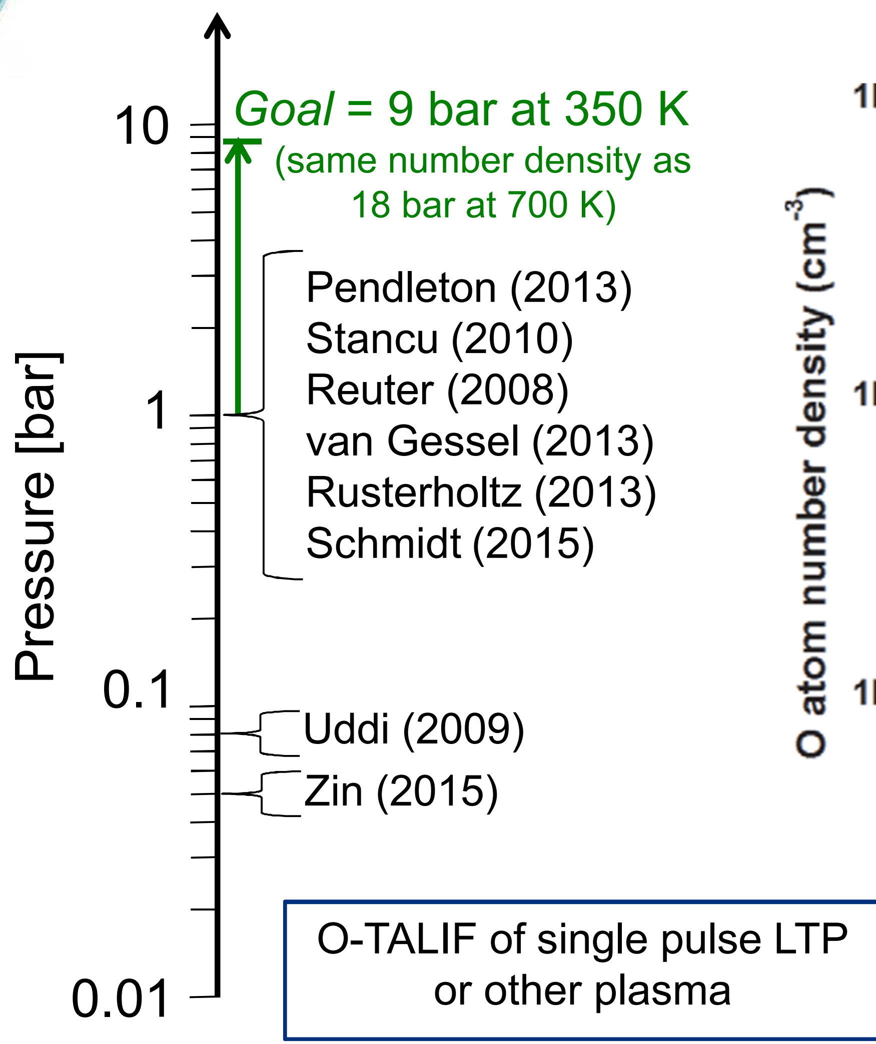
G.D. Stancu et al, *J. Phys. D: Appl. Phys.* 43 (2010) 124002

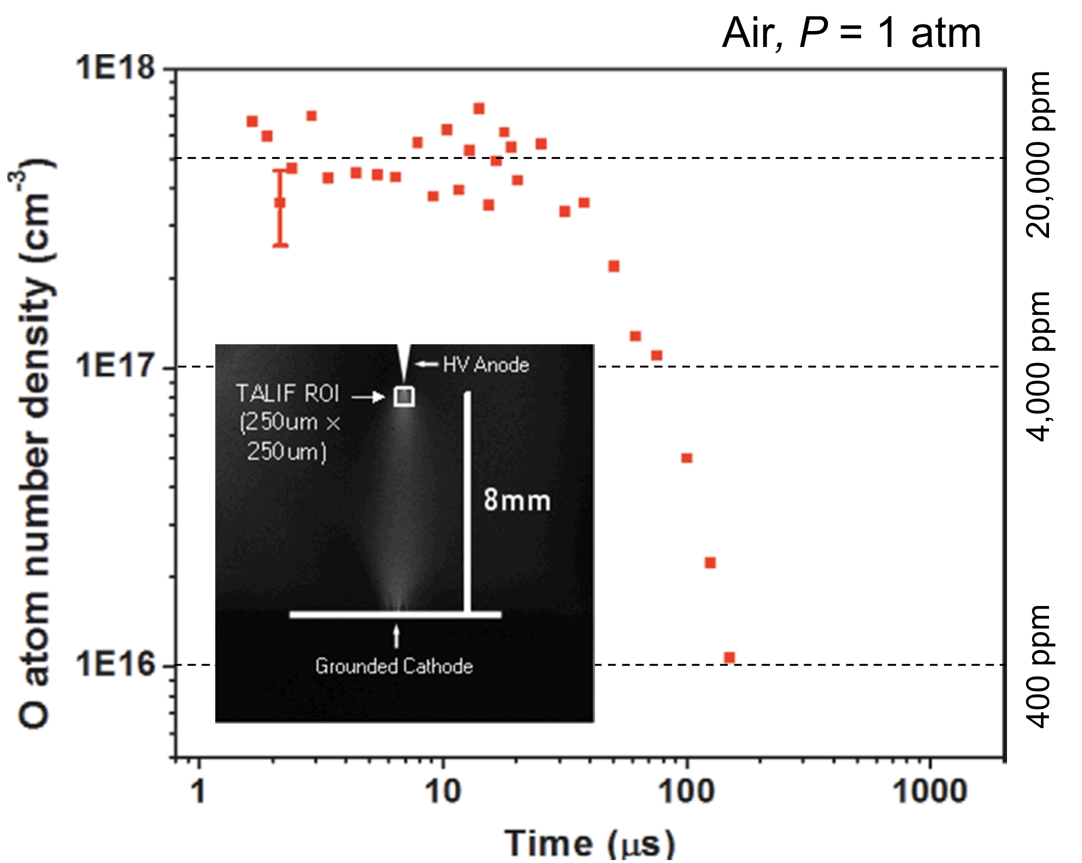
Correction factor

$$N_O = \frac{S_O}{S_{Xe}} g_{ND} \left(\frac{a_{21}(Xe)}{a_{21}(O)}\right) \left(\frac{\upsilon_O}{\upsilon_{Xe}}\right)^2 \frac{1}{F_O(T)} N_{Xe}$$

- Signals normalized by laser energy squared
- Correction for ND filter, quantum efficiency, BP filter
- Correction for photon count at same energy
- Boltzmann factor for lower level of oxygen ground state
- Fluorescence quantum yield $a_{21} = \frac{A_{21}}{A_{21} + Q}$

Existing O-TALIF measurements at or below atmospheric pressure; single shot LTP O-atom concentration profile as function of time





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