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Sandia National Laboratories

Imaging of Hydrogen Peroxide and Methyl in Nanosecond Pulsed Plasmas by Photofragmentation Laser-Induced Fluorescence

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Need a technique for spatially and temporally resolved imaging measurements of hydrogen peroxide (H_2O_2) and methyl (CH_3)

Key reactive species in surface decontamination, biomedical treatments, combustion, catalysis

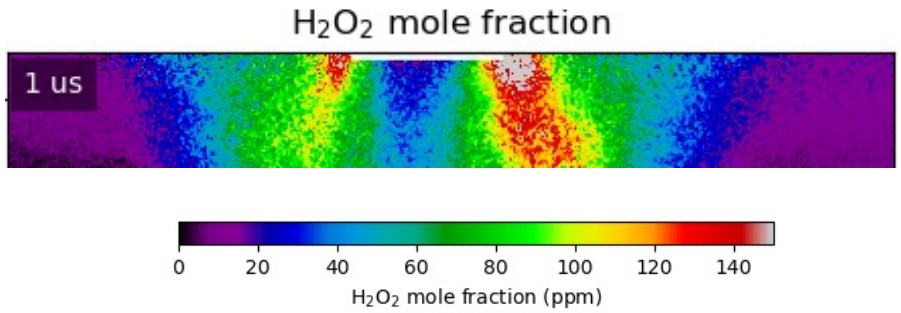
- Single-point and line-of-sight measurements
  - Molecular beam mass spectrometry (MBMS)
  - Absorption
  - Degenerate four-wave mixing (DFWM)
  - Resonance-enhanced multi-photon ionization (REMPI)

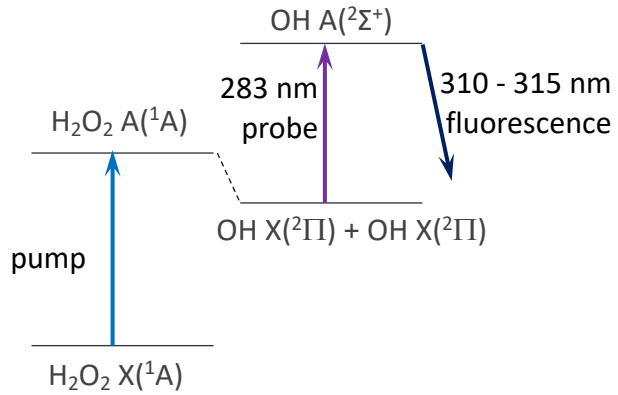
How to image hydrogen peroxide and methyl?

- H_2O_2 and CH_3 not directly detectable with laser-induced fluorescence (LIF) due to strongly predissociative excited states
- Detect OH product of photodissociation with OH-LIF: $H_2O_2 + hv \rightarrow 2OH$
- Detect CH product of photodissociation with CH-LIF: $CH_3 + hv \rightarrow CH + H_2$
- Previously demonstrated in combustion [Li et al. Proc. Combust. Inst. 34, 3573 (2013); Li et al. Proc. Combust. Inst. 37, 4487 (2017)]

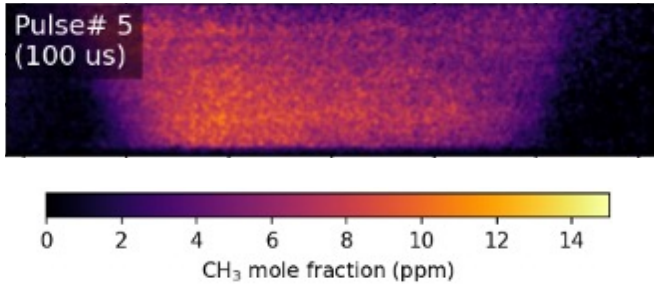
Photofragmentation Laser-Induced Fluorescence Enables Detection of Key Species that Don't Fluoresce

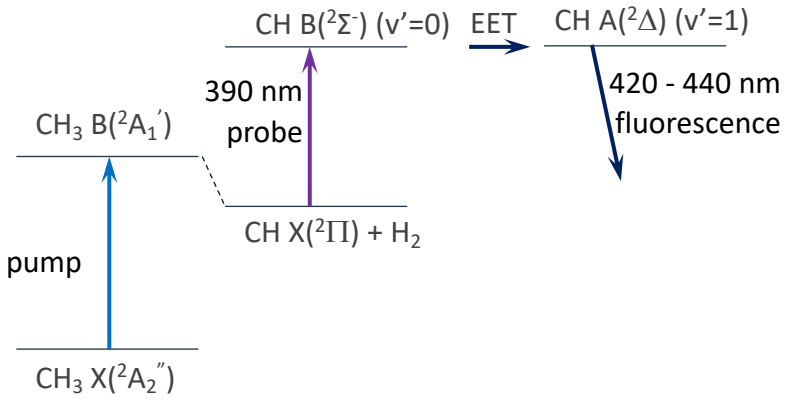
Hydrogen peroxide (H_2O_2)
Key species in interactions of plasma with water and tissue: decontamination/sterilization, plasma medicine



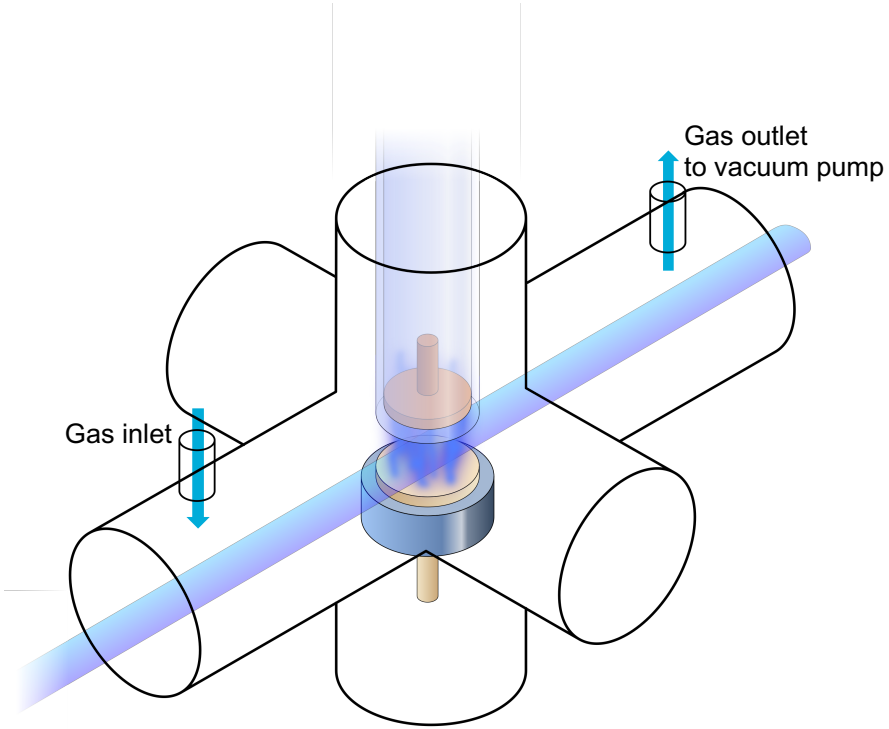


Methyl (CH_3)
Key intermediate for plasma-assisted catalysis, reforming, combustion

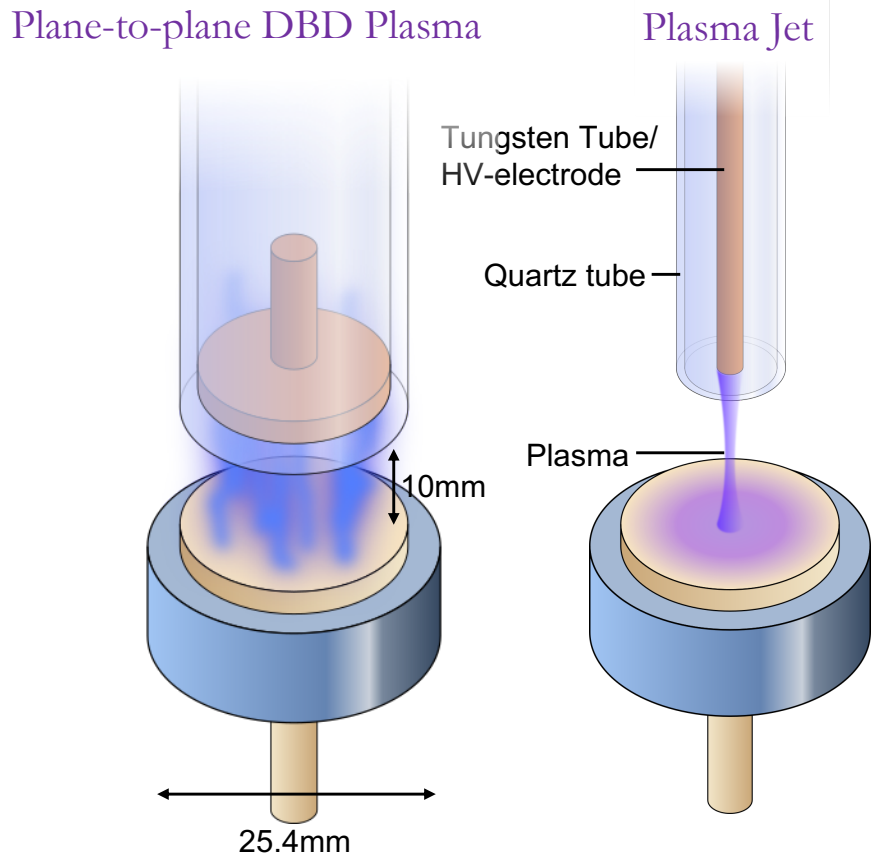




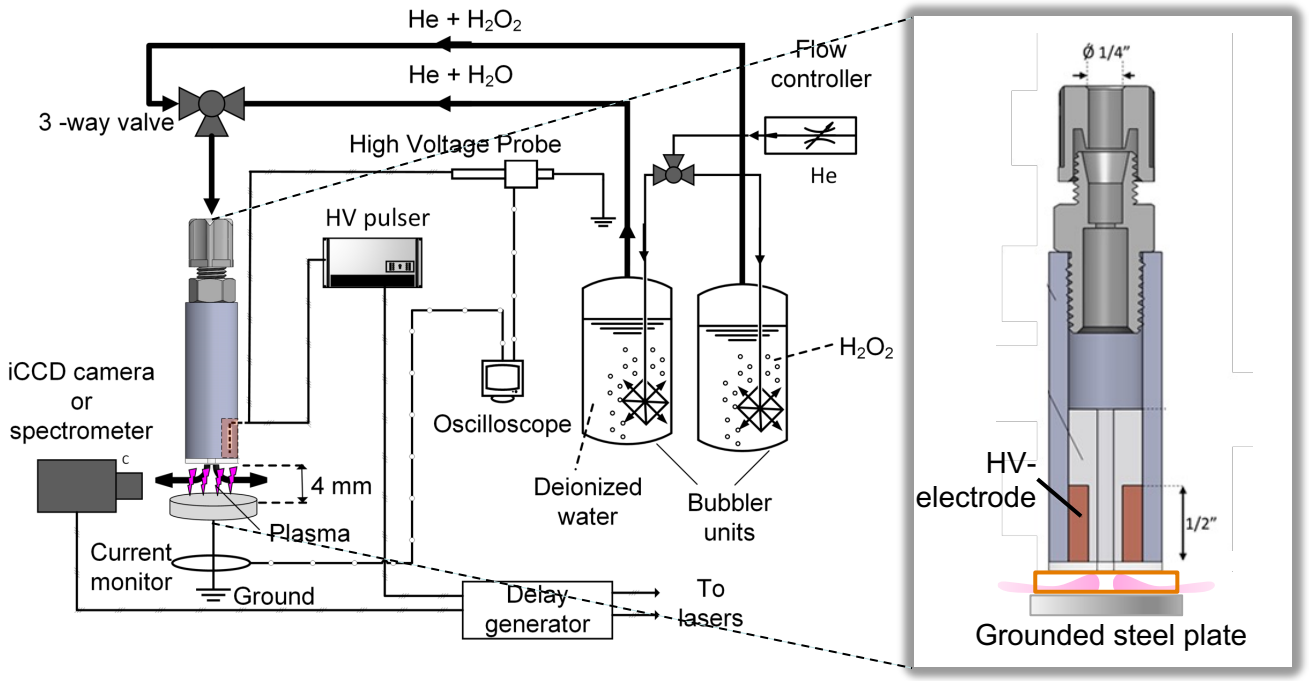
Configuration for 2D in-situ imaging of plasmas and plasma-surface interactions

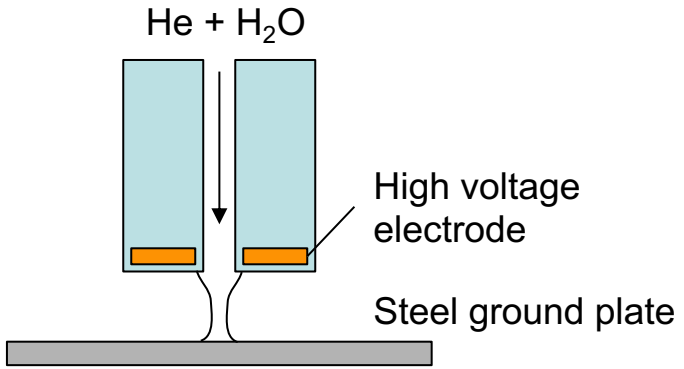


Our design: Direct plasma-catalyst interaction with optical access for in situ laser diagnostics



Quantification of Reactive Species and Characterization of High-Water Content in Dielectric Barrier Discharge using Photofragmentation and Laser-Induced Fluorescence



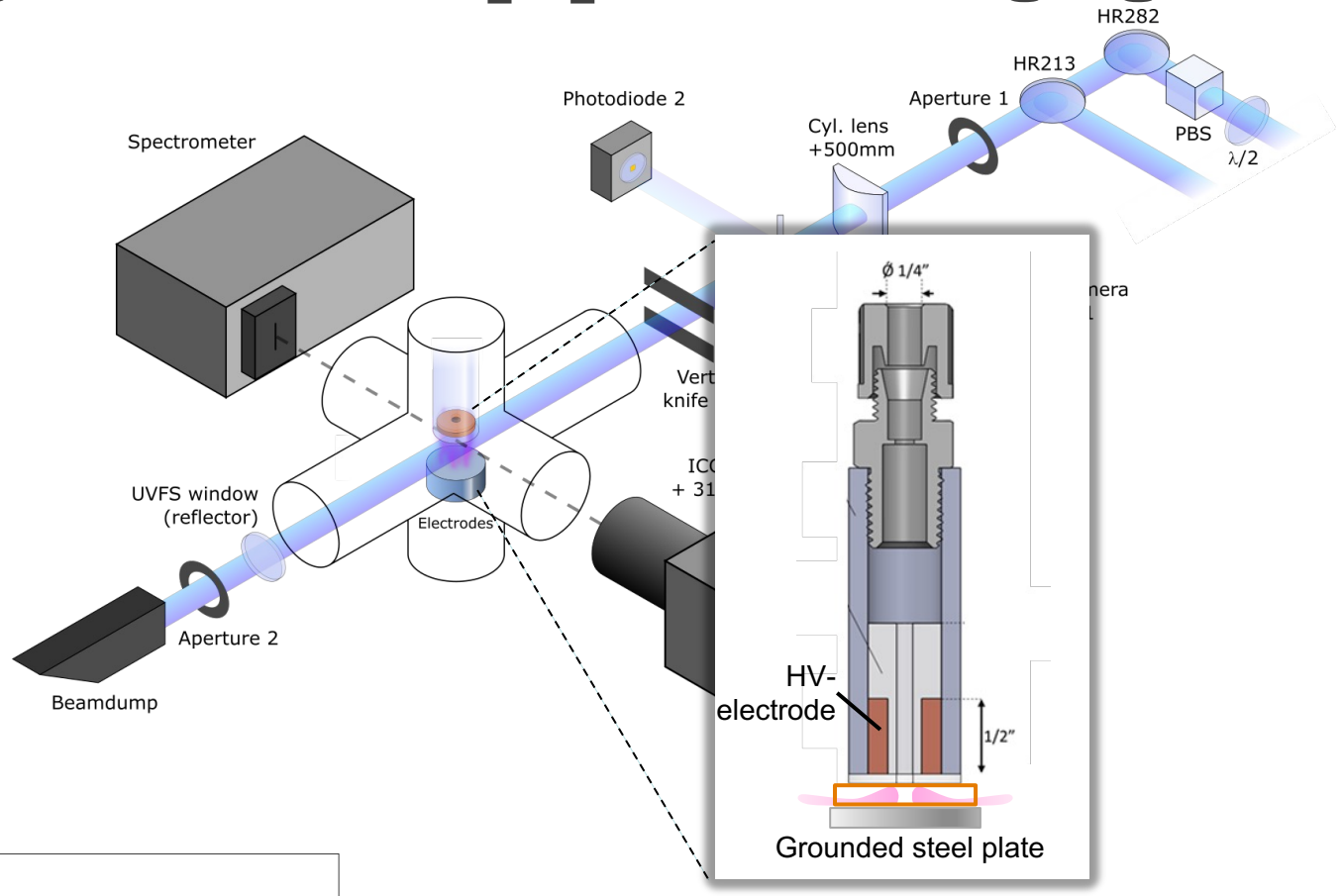


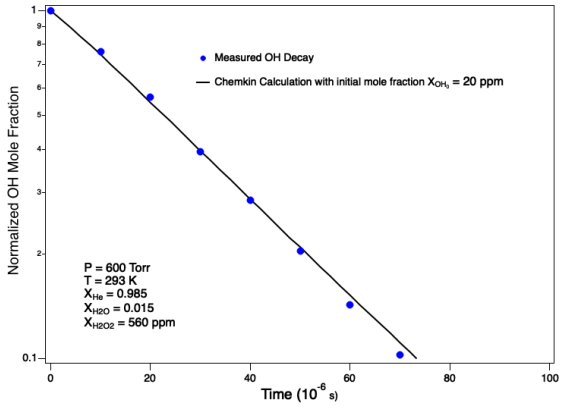
- Water saturated helium jet
- H_2O_2 PF-LIF ($H_2O_2 + h\nu_1 \rightarrow 2OH$)
- OH-LIF for measuring photolytically generated and plasma-generated OH

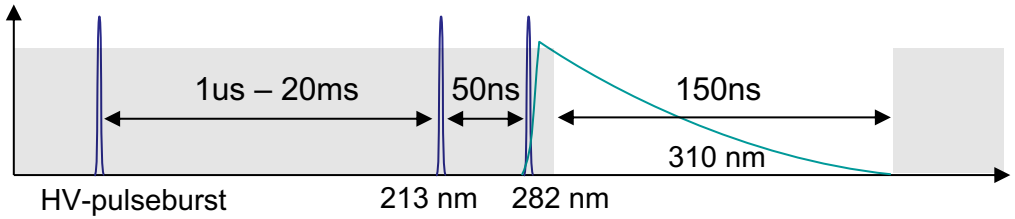
Measurements of key reactive species (OH, H_2O_2) needed for insights into properties of water-laden plasmas in decontamination, biomedical treatments

Experimental Configuration for H_2O_2 PF-LIF Imaging

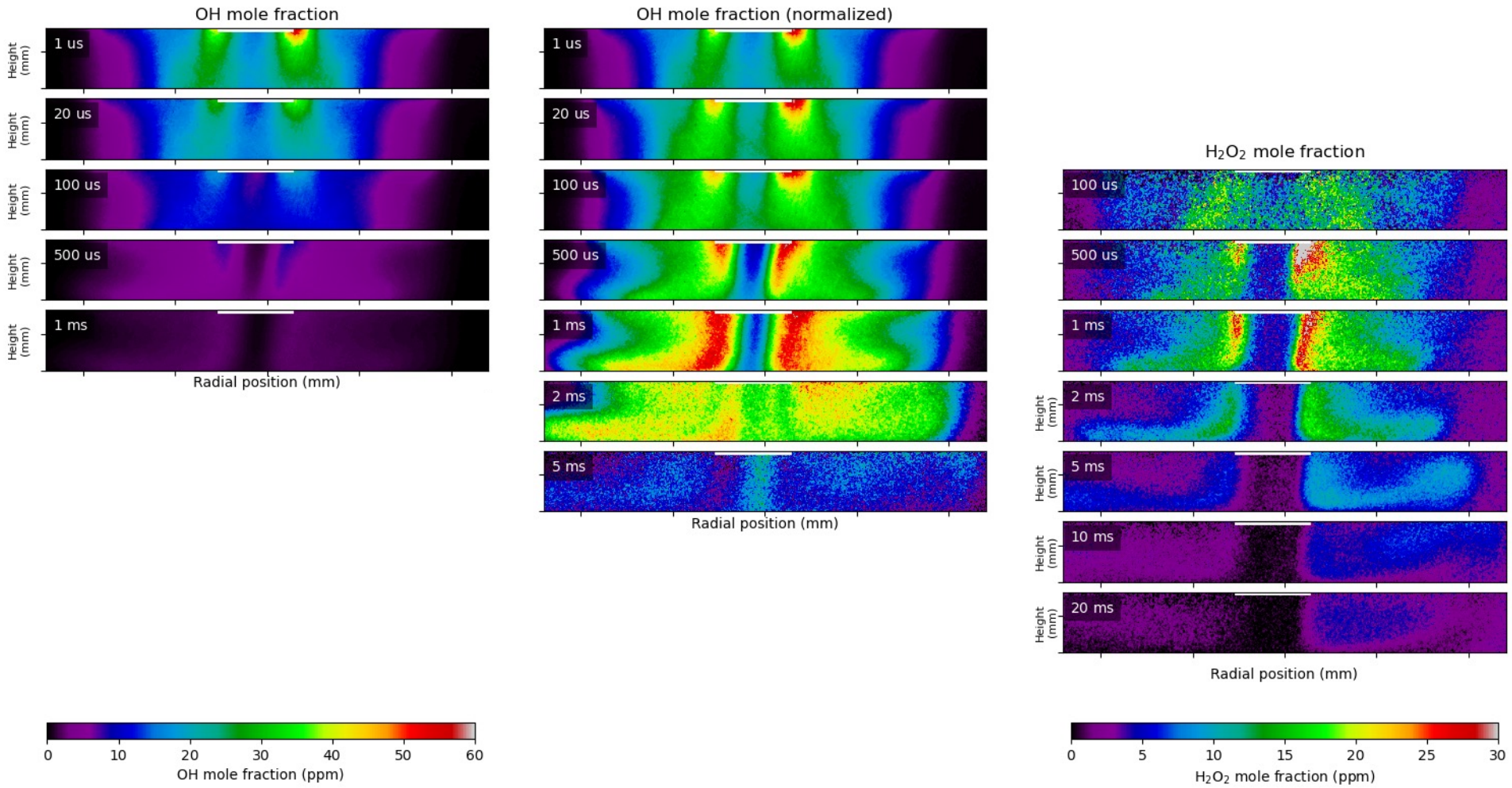
- PF-LIF lasers:
  - **PF:** 213 nm (5th HG YAG)
  - **LIF:** 282 nm (2nd HG 564nm dye)
- Conditions:
  - P = 600 torr
  - 4 slm He through H_2O bubbler (10°C)
  - 10kV, 5 pulse burst 50kHz/50Hz
- Calibration:
  - Reference mixture H_2O_2/H_2O/He
  - Reactive decay rate of OH compared to Chemkin simulations



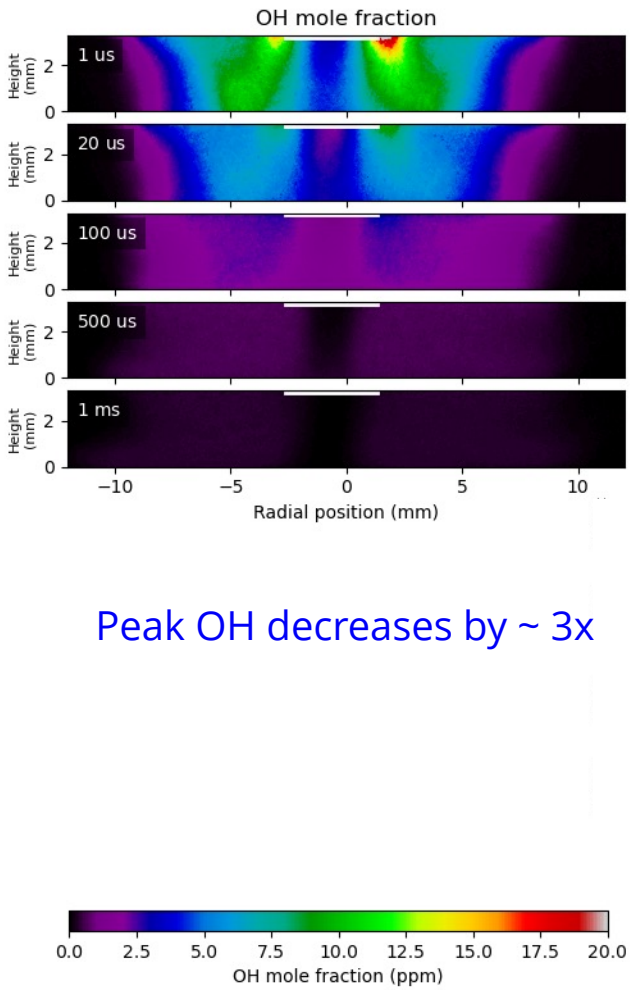




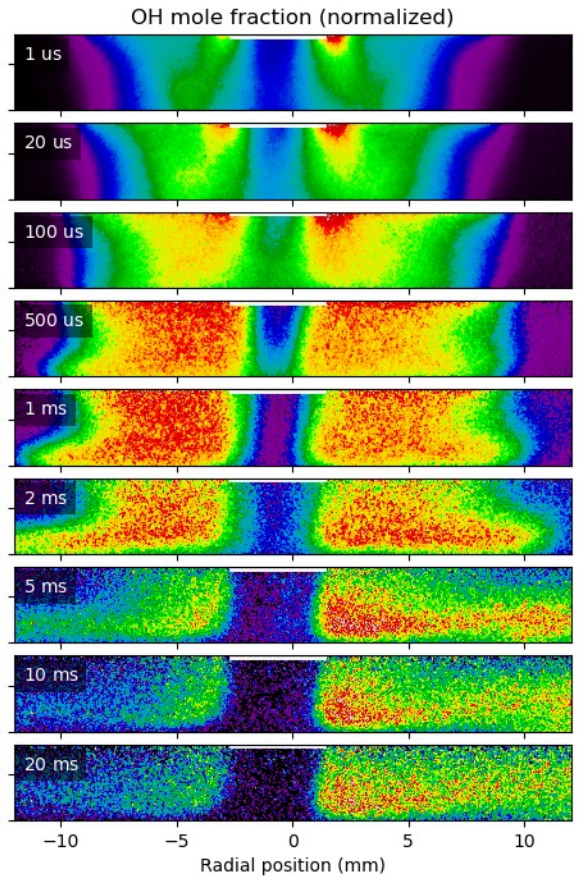
Imaging of H_2O_2 and OH in He-H_2O Jet

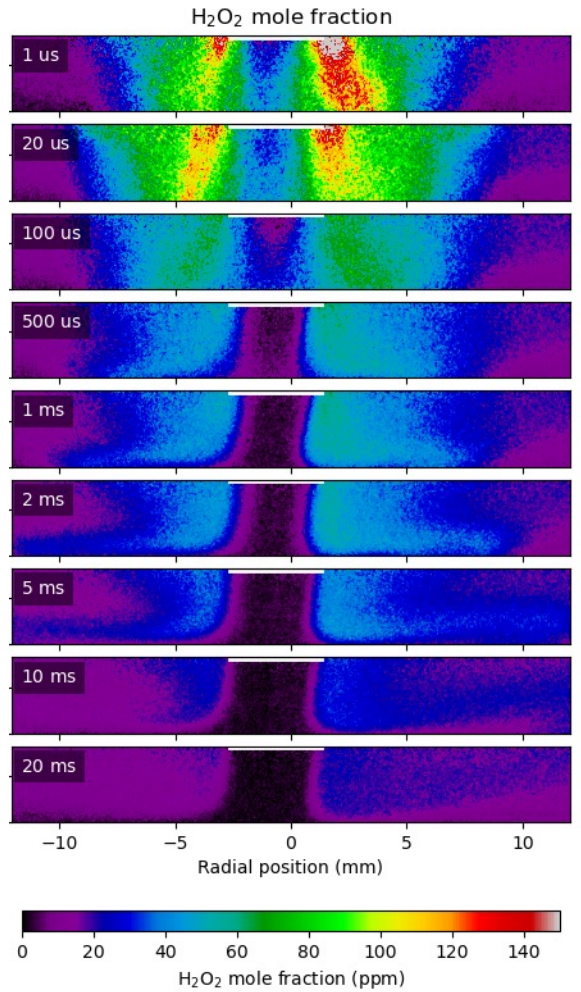


Imaging of H_2O_2 and OH in He-H_2O Jet with 5% O_2 Added



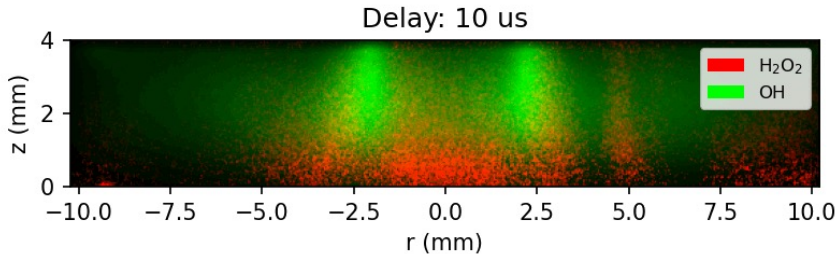
Peak OH decreases by ~ 3x





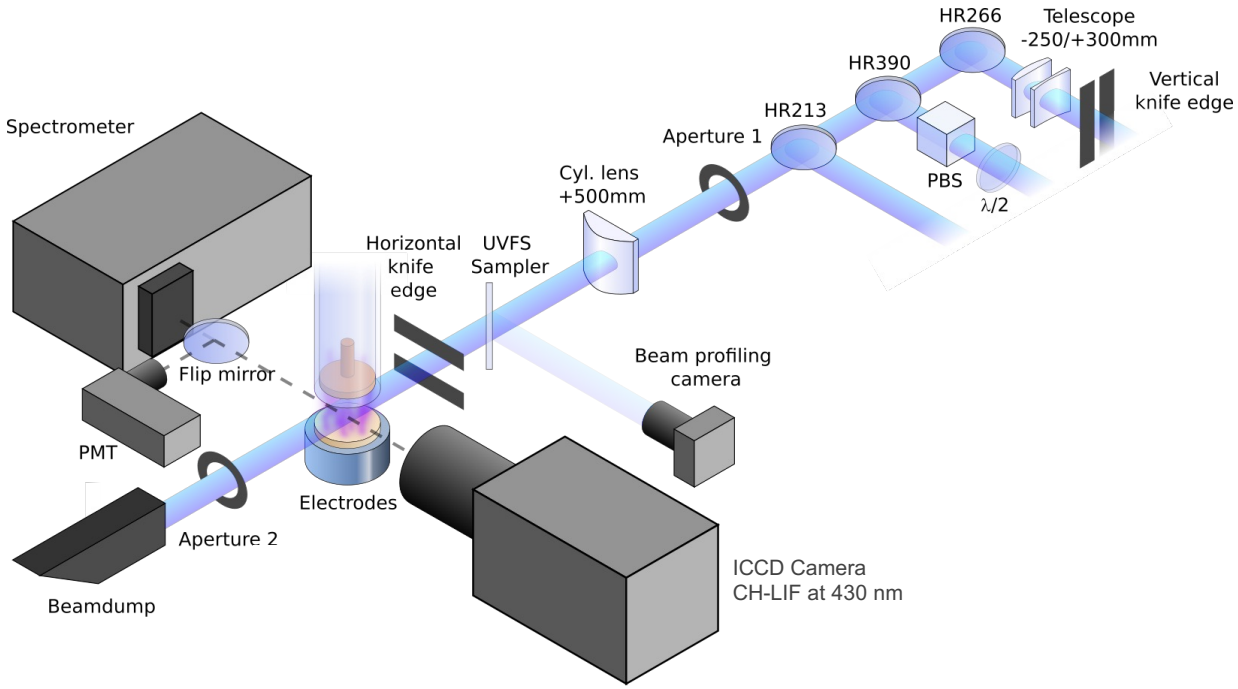
- Peak H_2O_2 increases by ~ 5x
- H_2O_2 production starts at earlier time

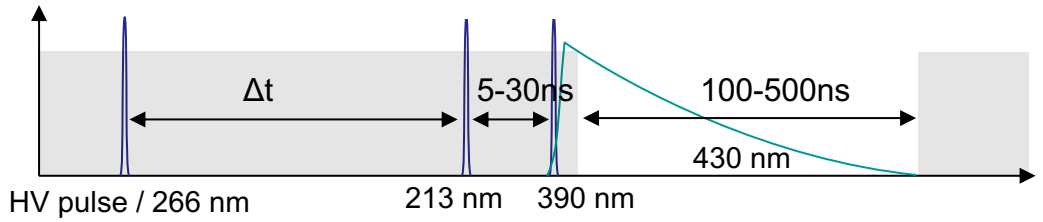
Summary H_2O_2/OH Imaging in Humid Plasma Jet



- Demonstrated quantitative 2D (PF)-LIF for imaging of OH and H_2O_2 distributions in a ns-plasma jet close to a surface
- Spatial and temporal evolution of H_2O_2 and OH show production, consumption, and transport of key reactive species near surface
- Effects of O_2 addition, flow rate, and pulse burst duration explored.
- Provides target case for modeling by Tanvir Farouk's group
- Enables further investigations of plasma-liquid/solid interactions

Configuration for Methyl Photofragmentation LIF

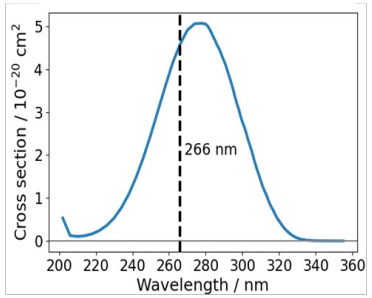




Methyl source for calibration:
Photodissociation of Acetone

$$CH_3COCH_3 + h\nu \rightarrow CH_3 + CH_3CO$$

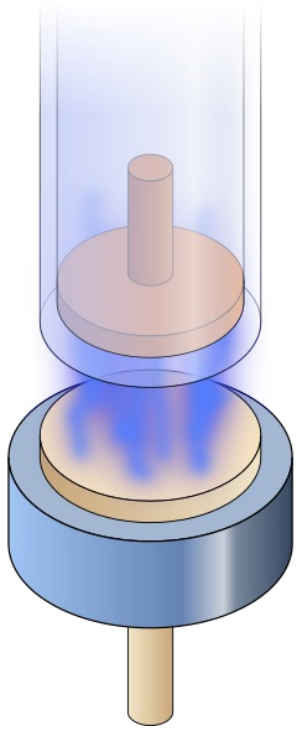
Acetone absorption

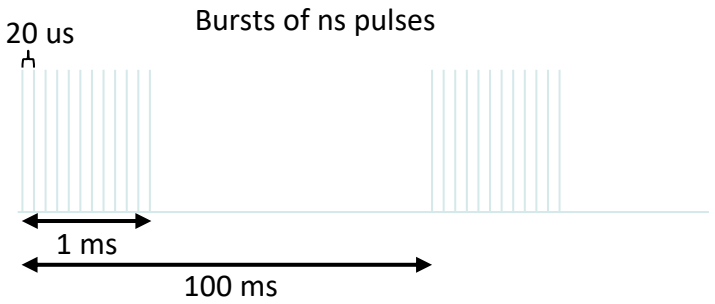


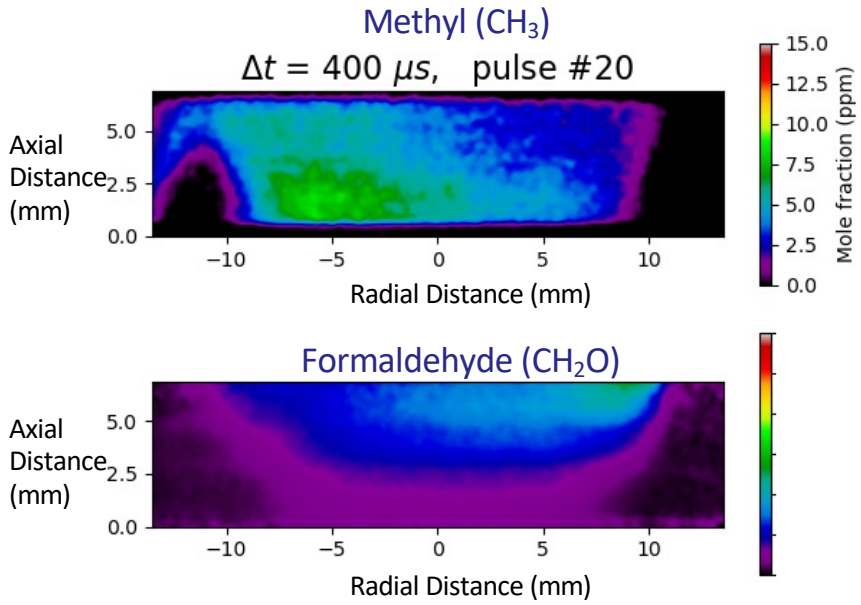
Acetone LIF used to measure temperature distribution and account for T dependence of absorption.

Temporal Evolution of Methyl and Formaldehyde in Plane-to-Plane Pulsed Dielectric Barrier Discharge Plasma

Plasma conditions:
- 1 ms burst of 16kV ns pulses @ 50 kHz
- 200 Torr
- 40% CH_4 , 40% CO_2, 20% He
- Lower surface: V_2O_5 Catalyst @ 200 C





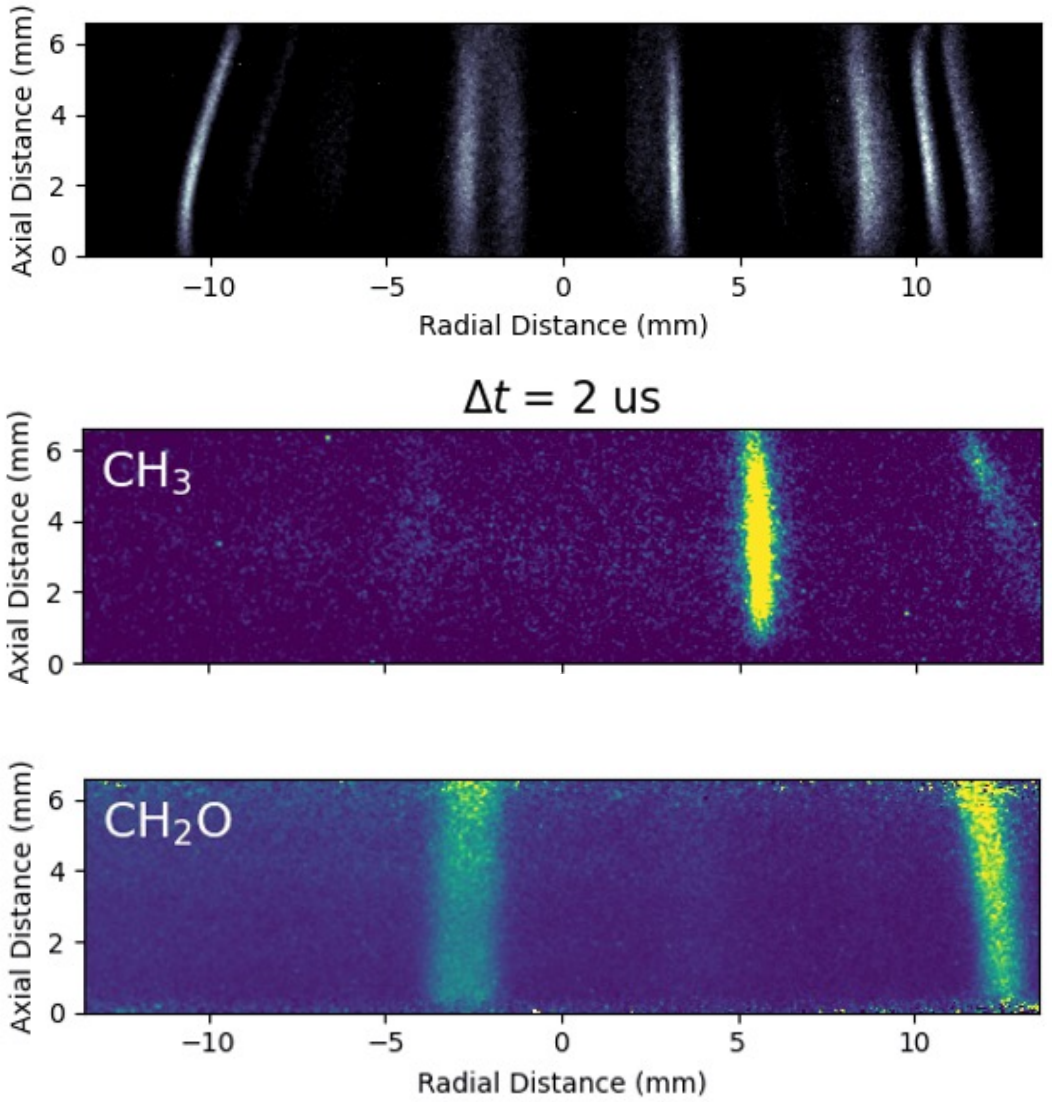


With Subtraction of Residual Signals from Previous Burst

Methyl and Formaldehyde Imaging in a Filamentary Discharge

Plasma conditions:

- 1 ms burst of 16kV ns pulses @ 50 kHz
- 100 Torr
- 83% CH_4 , 17% O_2
- Lower surface: V_2O_5



Summary CH_3 Imaging in Plasmas

- Demonstrated spatially and temporally resolved 2D imaging of methyl profiles in a plasma
- Near-surface detection using non-resonant LIF scheme
- Methyl PF-LIF calibration by photo-dissociation of acetone
- Quenching dominated by methane doesn’t vary between calibration and experiment
- Acetone LIF used for temperature corrections
- Absolutely calibrated time resolved 2D-methyl profiles were measured in a ns-plasma, obtaining
- Current collaboration with U Mich.: Comparisons of measurements and modelling of methyl production in nanosecond pulsed plasma
- Sets stage for studying plasma-assisted catalysis and pyrolysis

Acknowledgements

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