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Nonequilibrium Thermodynamics Laboratories

Fundamental Mechanisms, Predictive Modeling, and Novel Aerospace Applications of Plasma Assisted Combustion

Overview of OSU research plan

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**MURI Kick-Off Meeting
November 4, 2009**

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Thrust 1. Experimental studies of nonequilibrium air-fuel plasma kinetics using advanced non-intrusive diagnostics

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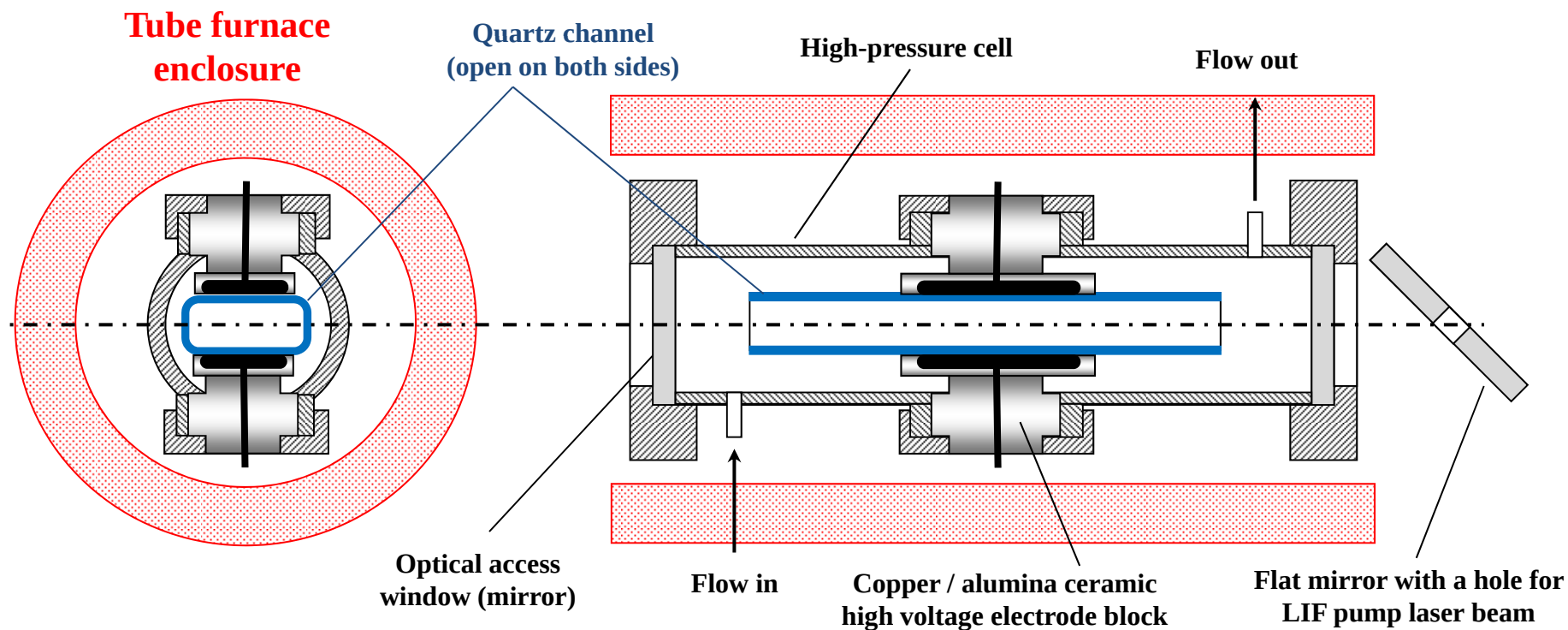
Task 1: Low-to-Moderate ($T=300-800$ K) temperature, spatial and time-dependent radical species concentration and temperature measurements in nanosecond pulse plasmas in a variety of fuel-air mixtures pressures ($P=0.1 - 5$ atm), and equivalence ratios ($\phi \sim 0.1-3.0$)

Goal: Generate an extensive set of experimental data on radical species concentrations and temperature rise; elucidate kinetic mechanisms of low-temperature plasma chemical fuel oxidation and ignition using kinetic modeling. Bridge the gap between room-temperature data (low-pressure gas discharges) and high-temperature data (shock tubes)

Test Bed #1: High-temperature, high-pressure nanosecond pulse discharge cell

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High-pressure discharge cell inside a tube furnace (6 inch bore, up to $T=1200^{\circ}\text{C}$)

Premixed fuel-air flow ($\sim 1\text{ m/s}$), preheated in the furnace, from 0.1 atm to a few atm

Repetitive nanosecond pulse discharge plasma: 20-40 kV, 5-25 nsec, 10 Hz to 100 kHz

Optical access (LIF, TALIF, CARS, CRDS) on the sides

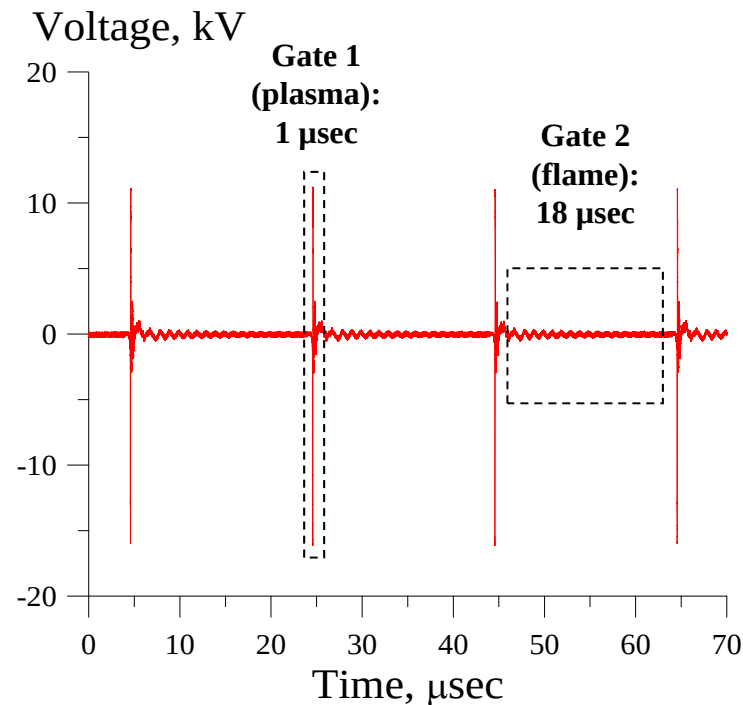
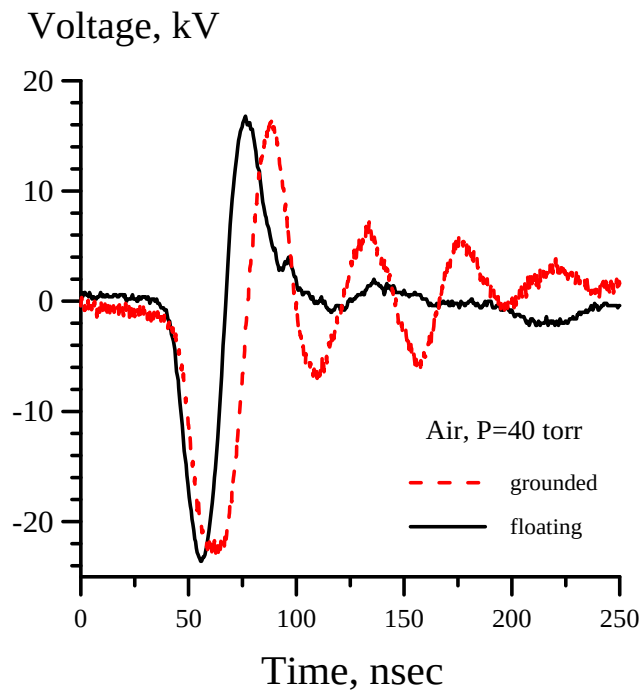
Fuels: hydrogen, methane, ethylene, propane, pentane, methanol & ethanol vapor

Repetitive nanosecond pulse plasma for kinetic studies:

Air, $P=60$ torr, $\nu=40$ kHz, 40 msec burst, 1 μ sec gate

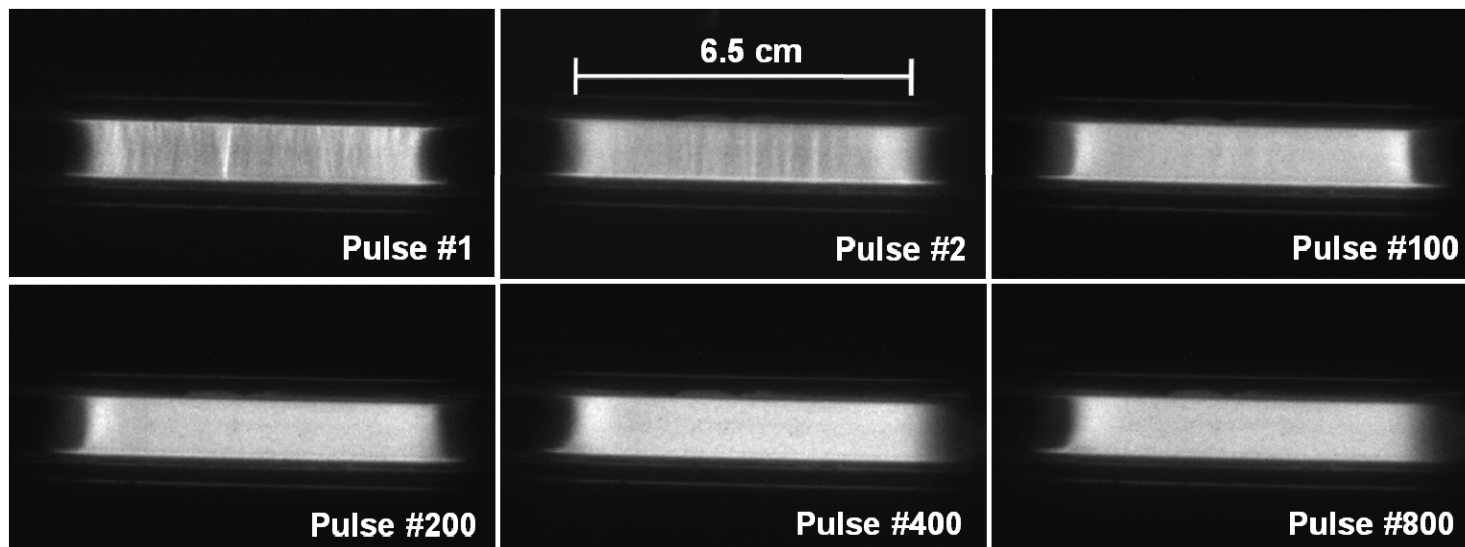
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- Some filamentary structure in pulses #1 and #2

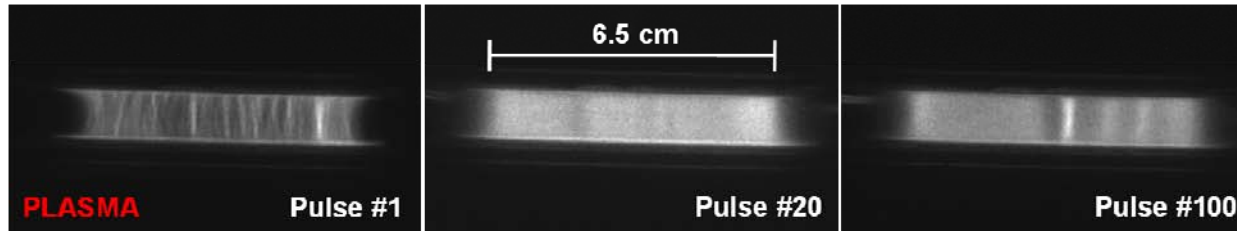
- Uniform air plasma during subsequent pulses, at $P=40$ -100 torr



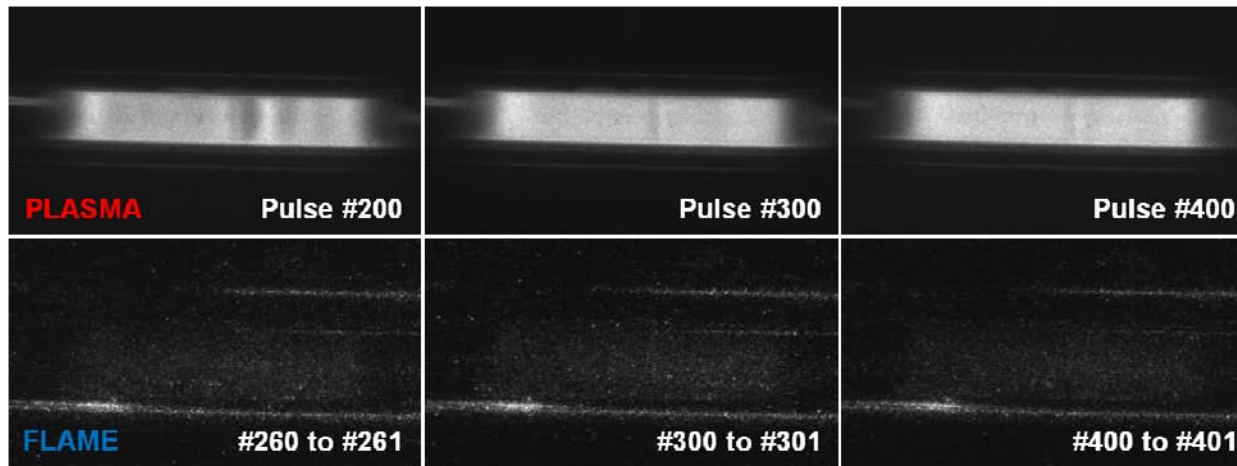
Repetitive nanosecond pulse plasma for kinetic studies: Ethylene-air, $P=40$ torr, $\phi=1$, $\nu=40$ kHz

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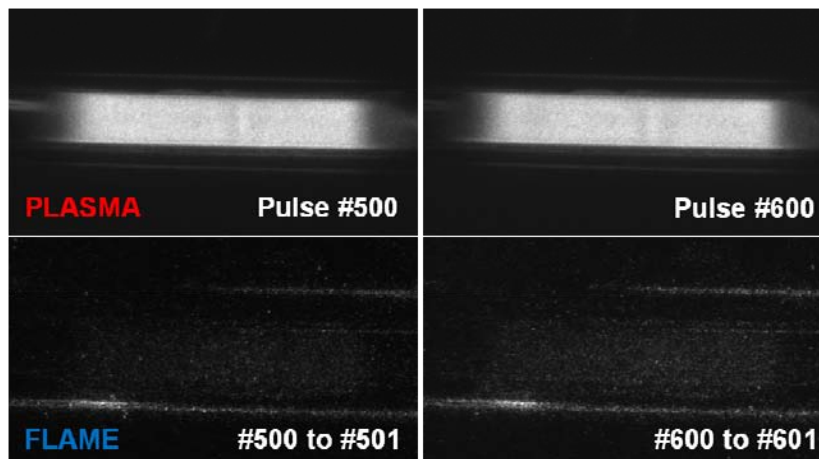


- Nearly uniform plasma during entire burst (except pulses #1 and #2)



- Ignition does not occur, likely due to rapid wall cooling

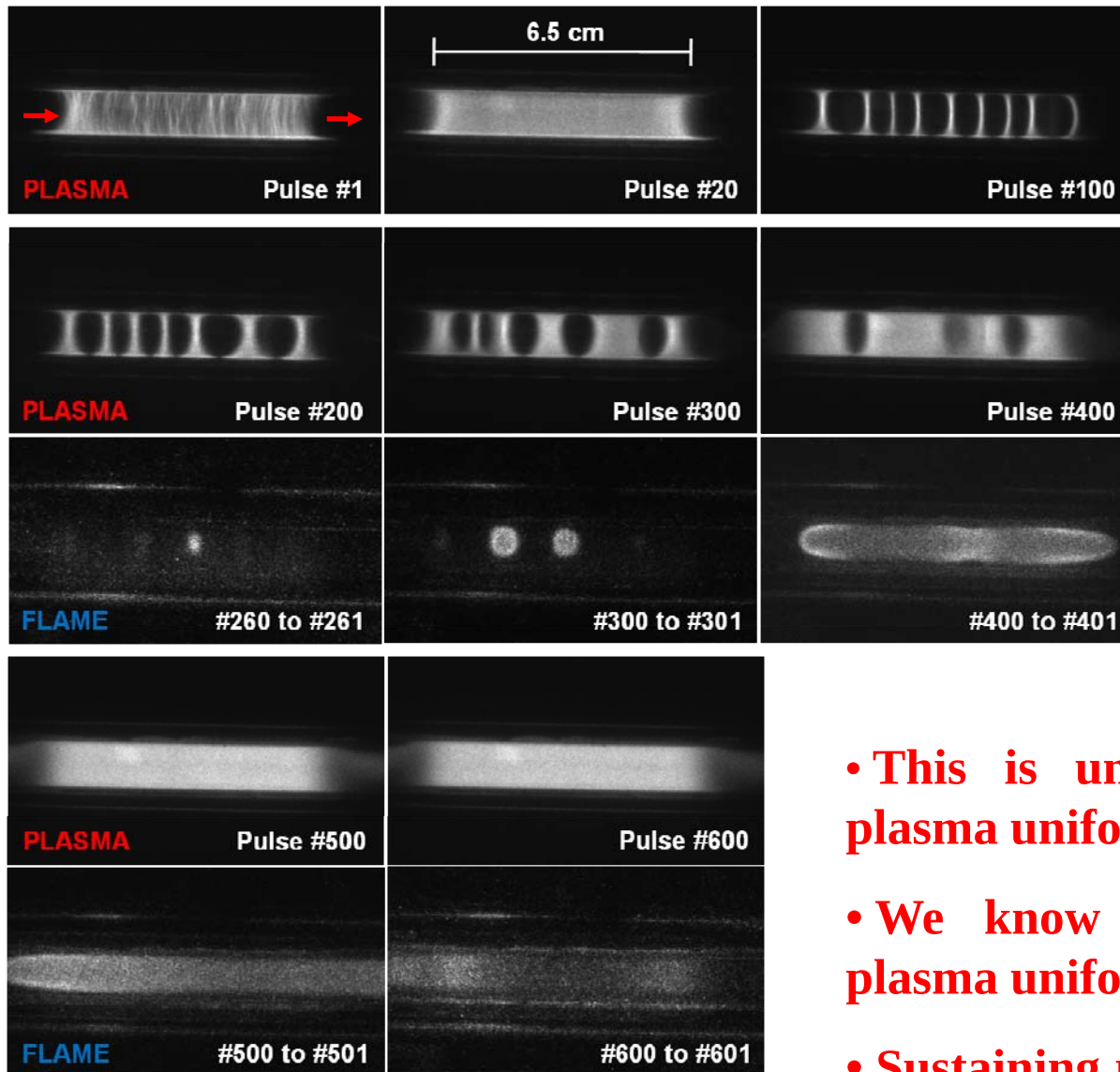
- Pressure is low – can this experiment be done at higher pressures?



Repetitive nanosecond pulse plasma for kinetic studies: Ethylene-air, $P=60$ torr, $\phi=1$, $\nu=40$ kHz

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- Uniform plasma during first few tens of pulses (except pulses #1 and #2)

- Well-defined filaments form in pulse #100, persist for several hundred pulses

- After ignition occurs, flame fills entire discharge volume, and plasma becomes uniform again

- Filamentation likely due to ionization / heating instability

- This is unacceptable: need to keep the plasma uniform during entire burst

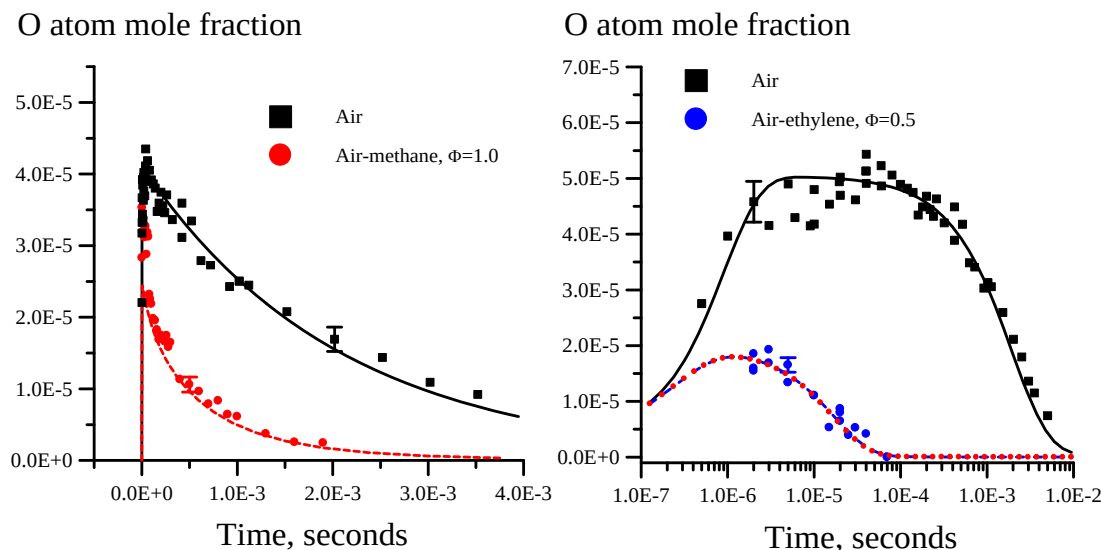
- We know that preheating will improve plasma uniformity

- Sustaining plasma in a heated cell will allow measurements at higher pressures

Time-resolved species concentrations: O and H atoms (Two-Photon Absorption LIF with Xe and Kr calibration)

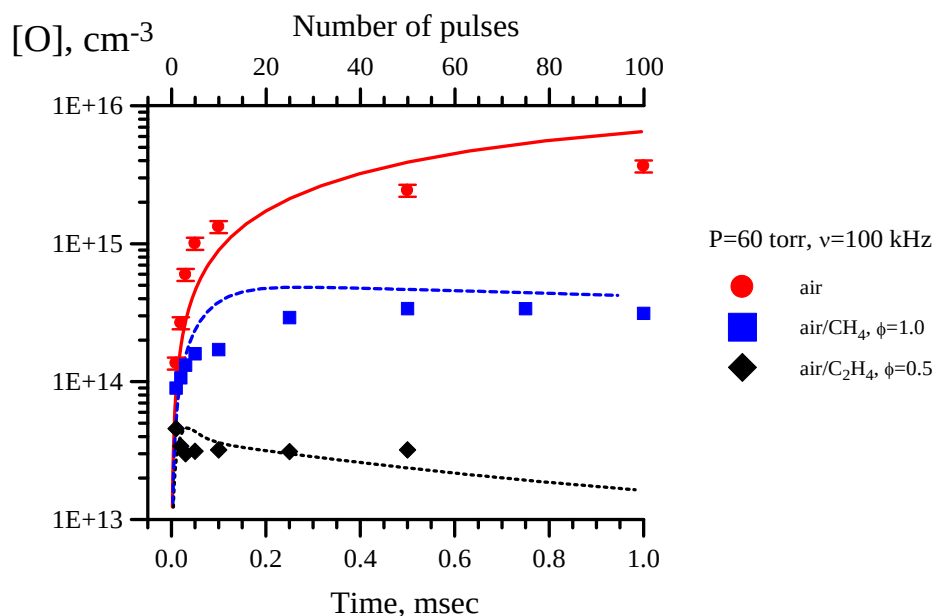
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Previous results: O atoms in air, methane-air, and ethylene-air at $P=60$ torr (single-pulse and burst mode, initially at $T=300$ K)

Objective: measure time-resolved O and H atoms in nsec pulse discharge plasmas in H_2 -air and C_xH_y air mixtures, at $P \sim 0.1 - 1$ atm, $T=300-800$ K

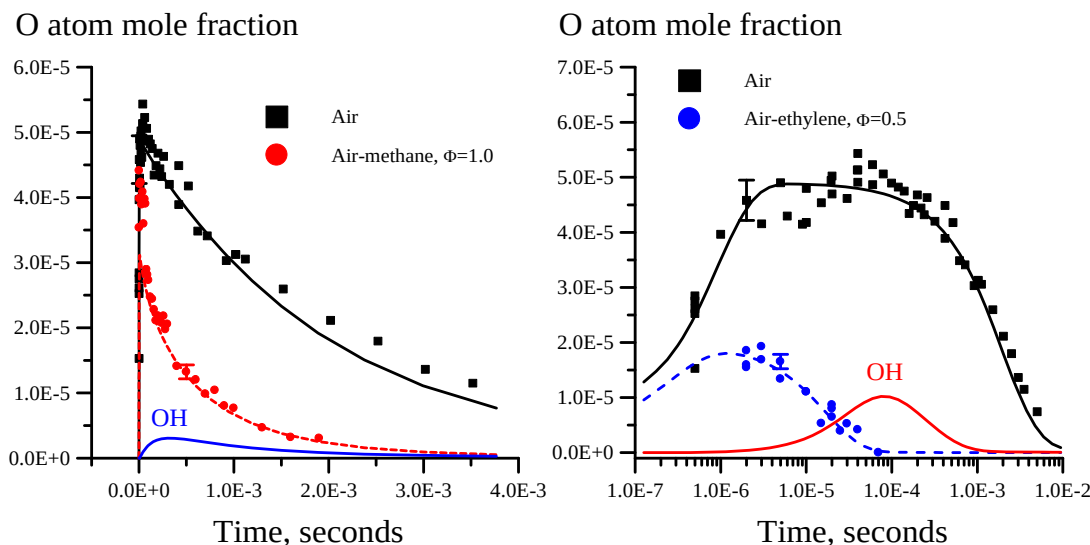


Outcome: kinetic mechanism of low-temperature plasma fuel dissociation and oxidation (specifically rates of O atom generation in the plasma and O atom reactions with fuel species)

Time-resolved species concentrations: OH (LIF with Hencken adiabatic burner calibration)

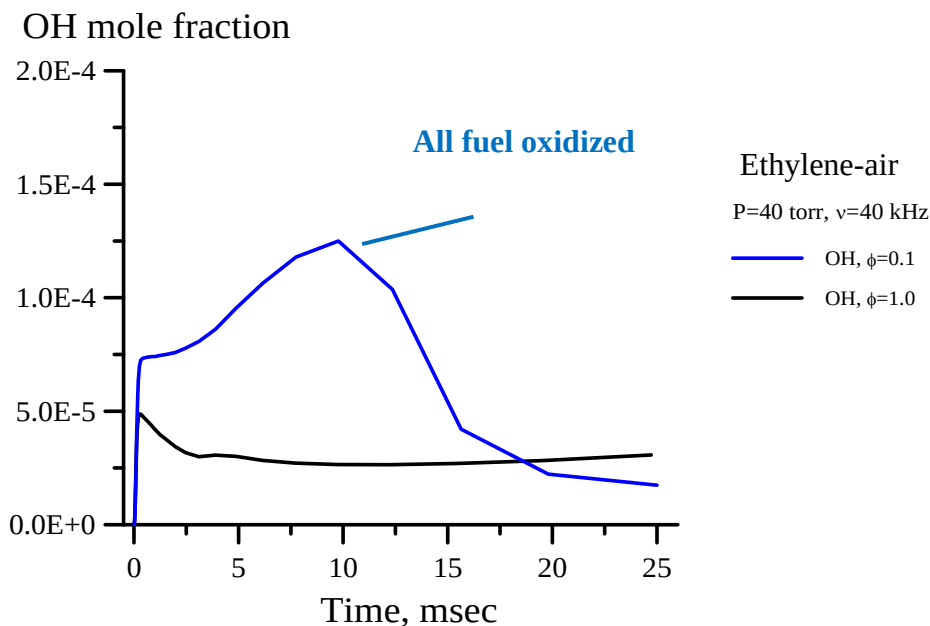
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Work currently underway: OH in methane-air and ethylene-air at $P=60$ torr (single-pulse and burst mode, initially at $T=300$ K)

Objective: measure time-resolved OH in nsec pulse discharge plasmas in H_2 -air and C_xH_y air mixtures, at $P \sim 0.1 - 1$ atm, $T=300-800$ K

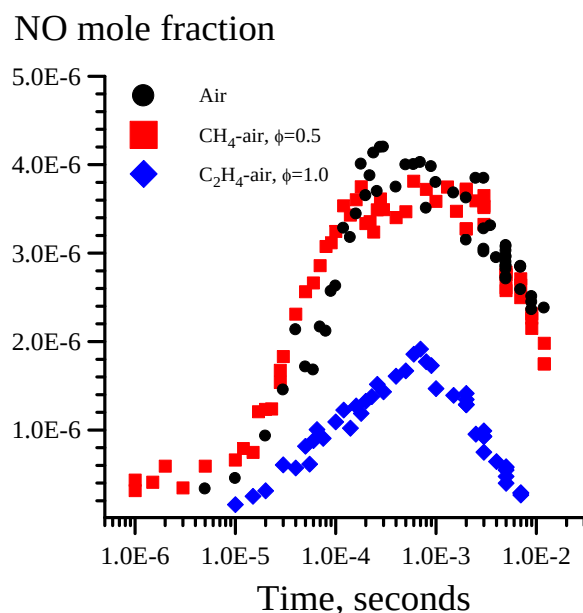
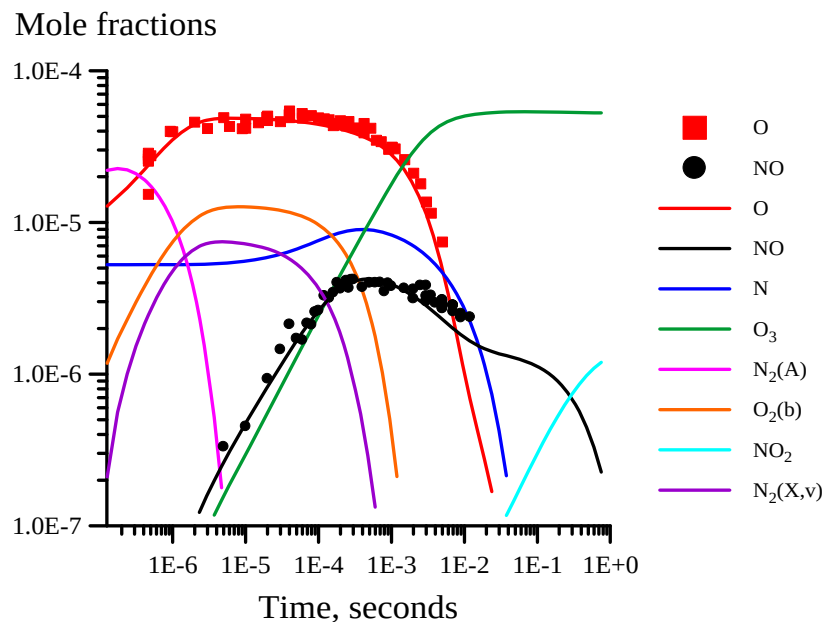


Outcome: kinetic mechanism of low-temperature plasma fuel oxidation (specifically rates of H atom abstraction from fuel species)

Time-resolved species concentrations: NO (LIF with calibration using known NO-N₂ mixture)

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Previous results : NO in air, methane-air and ethylene-air at $P=60$ torr (single-pulse, initially at $T=300$ K). State-of-the-art kinetic models cannot explain time-resolved data. Possible effect of $N_2(X,v) + O$ reaction.

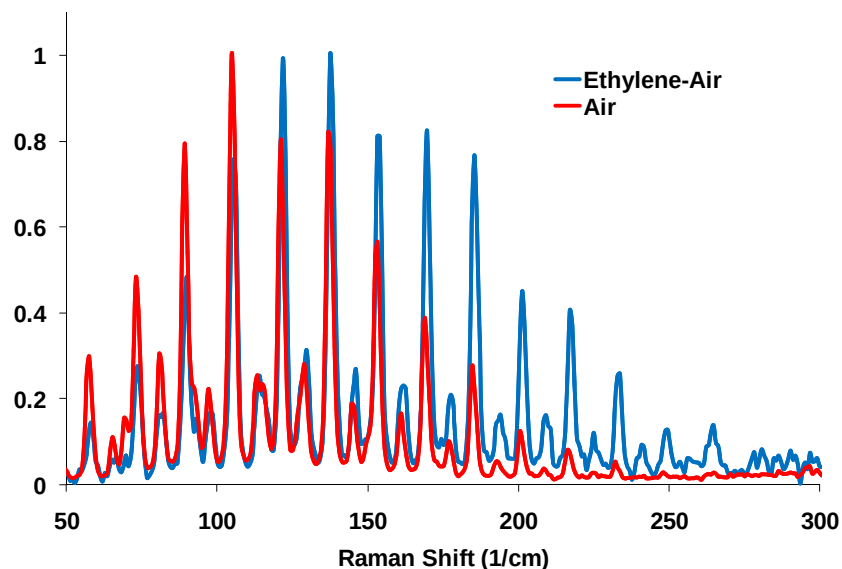
Objective: measure time-resolved NO in nsec pulse discharge plasmas in H₂-air and C_xH_y air mixtures, at $P \sim 0.1 - 1$ atm, $T=300-800$ K

Outcome: kinetic mechanism of low-temperature plasma fuel oxidation (specifically O₂ dissociation vs. NO formation in N₂* reactions)

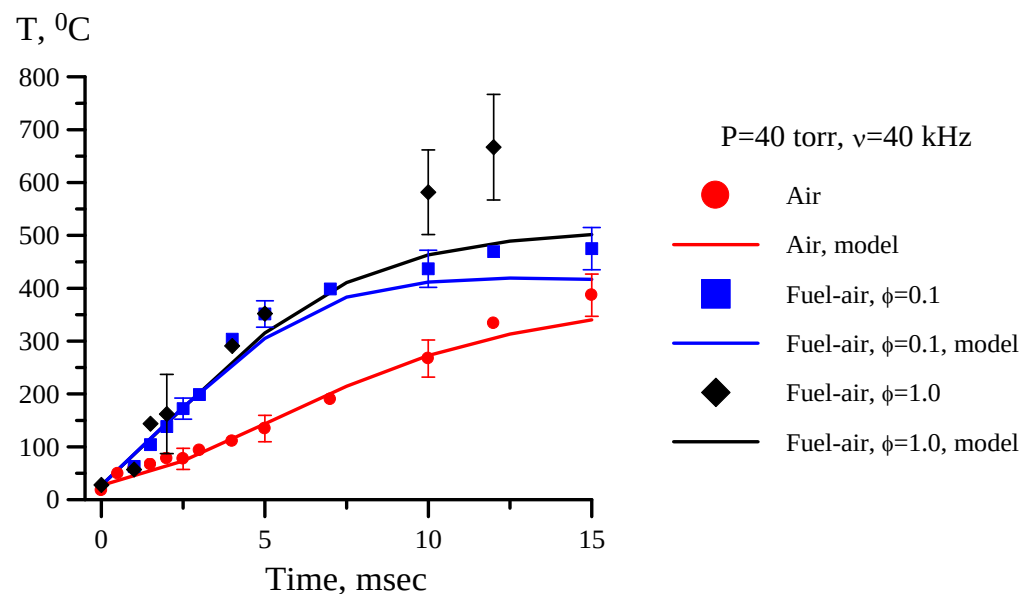
Time-resolved, spatially resolved temperature (purely rotational CARS)

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Previous results: time-resolved temperature in air and ethylene-air at $P=40$ torr (burst mode, initially at $T=300$ K). Evidence of significant additional heat release in fuel-air, compared to air



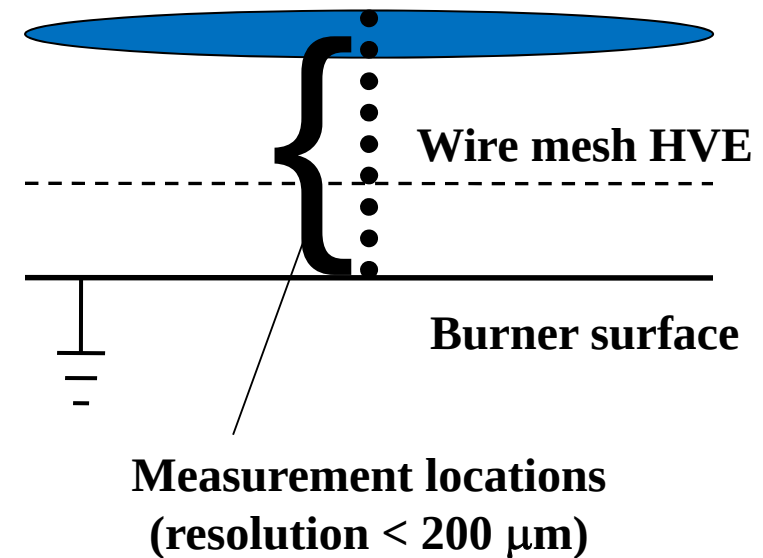
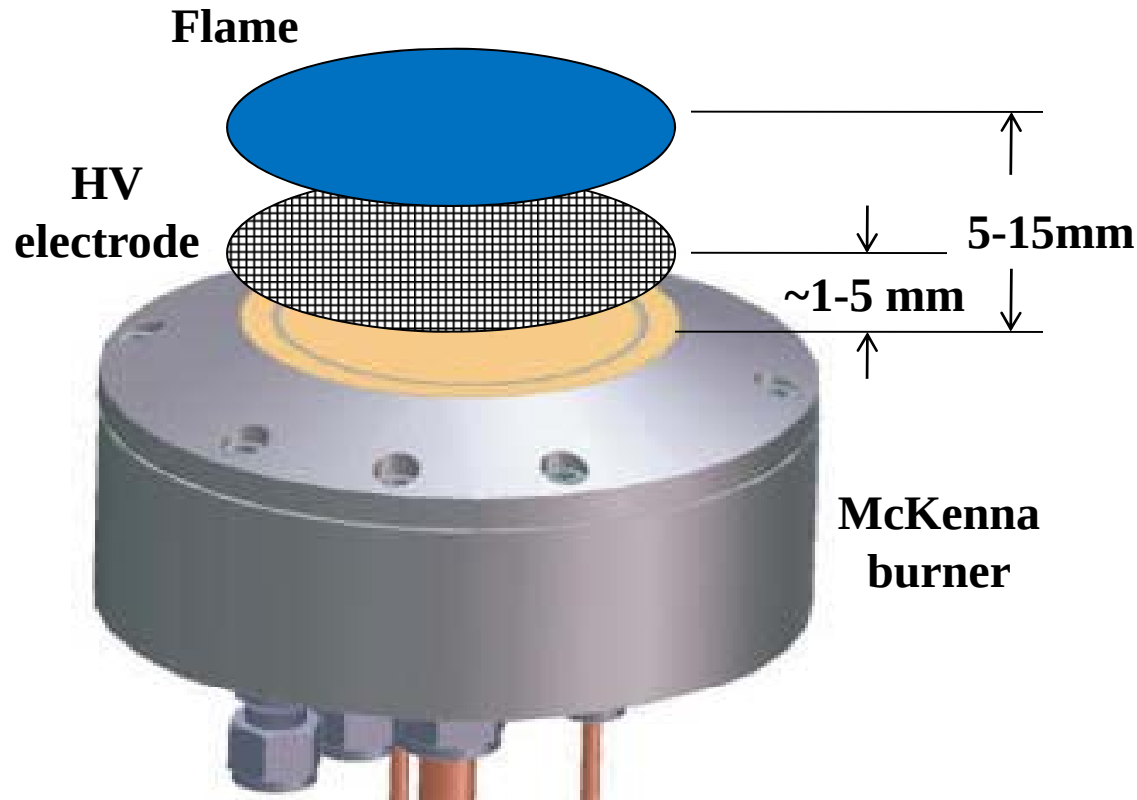
Objective: measure temperature in nsec pulse discharge plasmas in H_2 -air and C_xH_y air mixtures, at $P \sim 0.1 - 1$ atm, $T=300-800$ K

Outcome: kinetic mechanism of low-temperature plasma chemical energy release in exothermic fuel oxidation reactions with radicals

Test Bed #2: Flat flame McKenna burner with nanosecond pulse discharge

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Flat flame burner inside a six-arm cross vacuum chamber (8 inch bore)

Premixed fuel-air flow (~ 0.1 - 1.0 m/s) with N_2 co-flow, $P=10$ - 40 torr

Repetitive nanosecond pulse discharge plasma: 20 - 40 kV, 5 - 25 nsec, 10 Hz to 100 kHz

Optical access (LIF, TALIF, CRDS) on two perpendicular axes

Fuels: hydrogen, methane, ethylene, propane, pentane, methanol & ethanol vapor

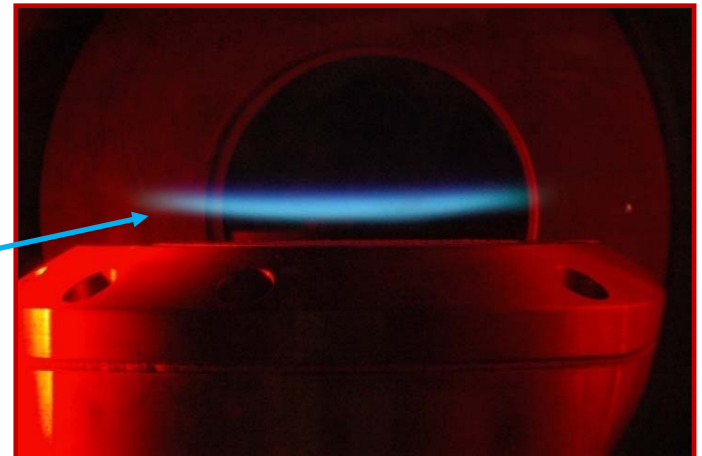
Interaction of plasma and flame chemistry: spatially resolved species concentrations and temperature

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- **Steady, laminar, low-pressure flat flames allow spatially-resolved measurements of temperature and species concentrations**
- **Minimize transport influence; isolate kinetic effects**
- **Can investigate full range of temperature conditions (from below 500 K to 2000 K) by adjusting measurement position (i.e. height above burner)**
- **Typical spatial scale ~5-20 mm, spatial resolution <200 μm**
- **Straightforward integration of nsec discharge plasma into a low-pressure flame facility and study of plasma effects (i.e. measurements with plasma “off” and “on”)**

Steady, laminar, 30 Torr, 1-D flame



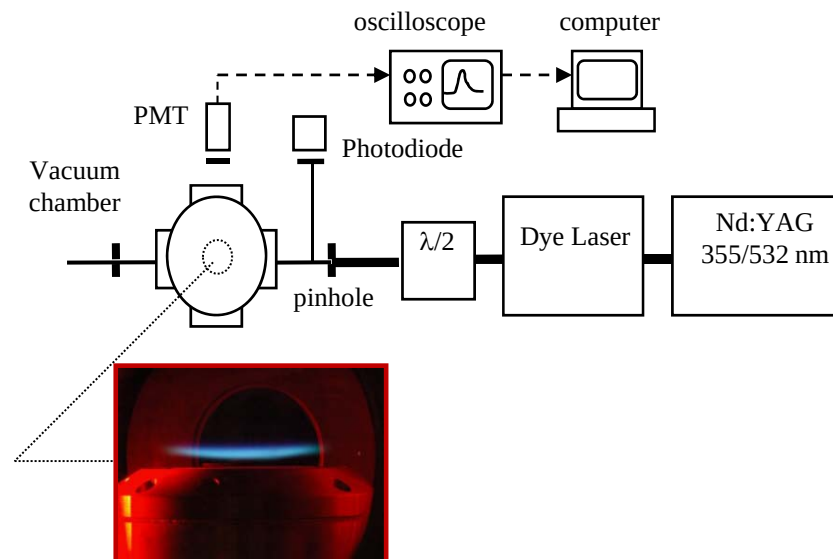
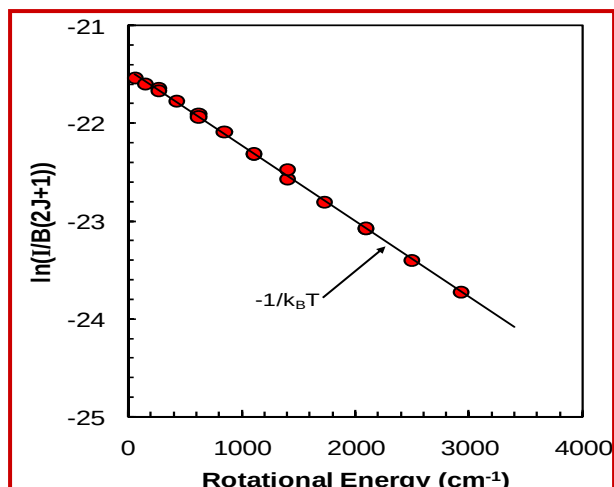
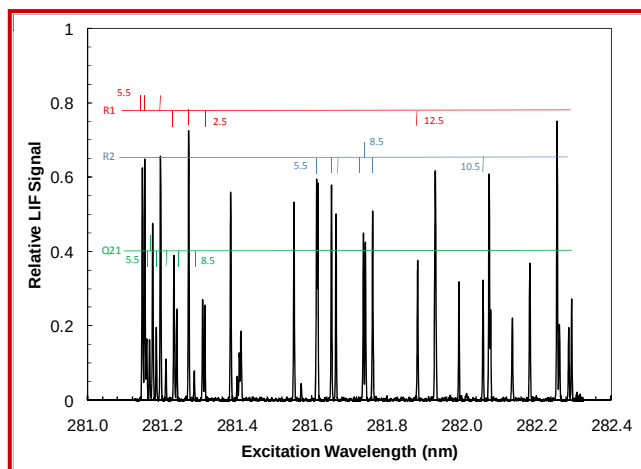
Previous low-pressure flame results (LIF):

P=10-40 torr; CH₄, C₂H₆, C₃H₈, C₄H₁₀; $\phi=0.6$ -1.4

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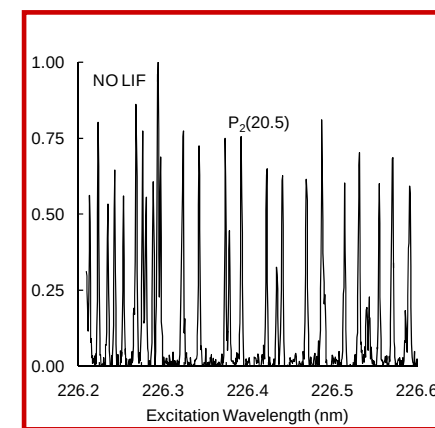
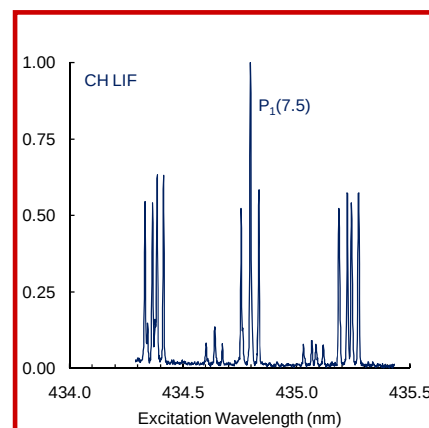
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Flame temperature from rotational structure of **OH A-X (1,0)** band near 282 nm



Spectral features used for profiles of flame species:

CH A-X (0,0) at 435 nm **NO A-X (0,0)** at 226 nm

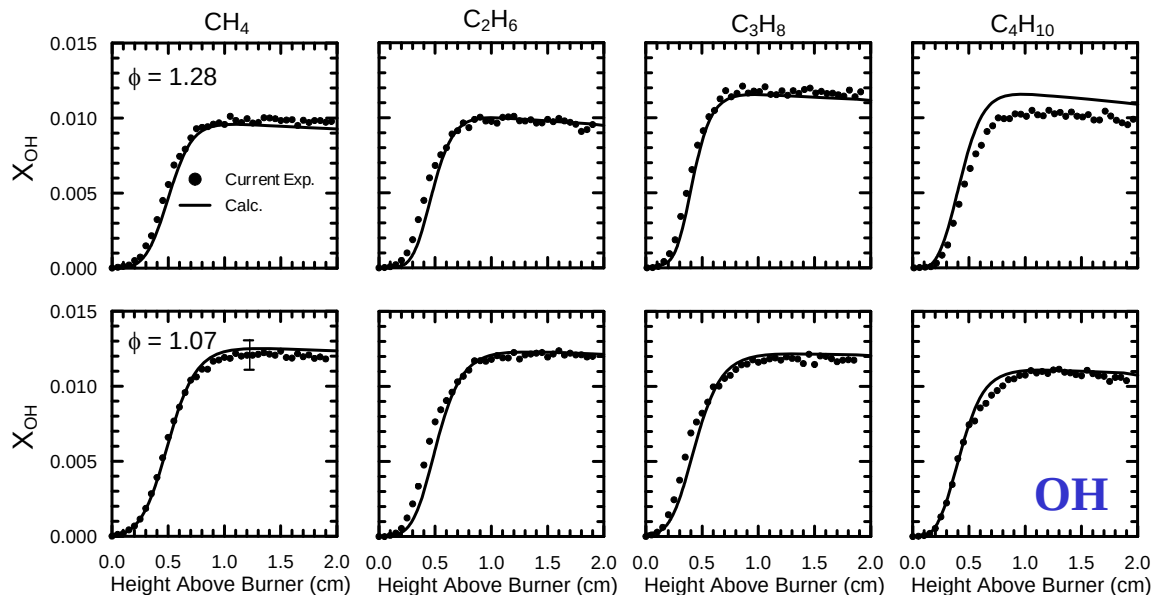


Previous low-pressure flame results (LIF):

P=10-40 torr; CH₄, C₂H₆, C₃H₈, C₄H₁₀; $\phi=0.6$ -1.4

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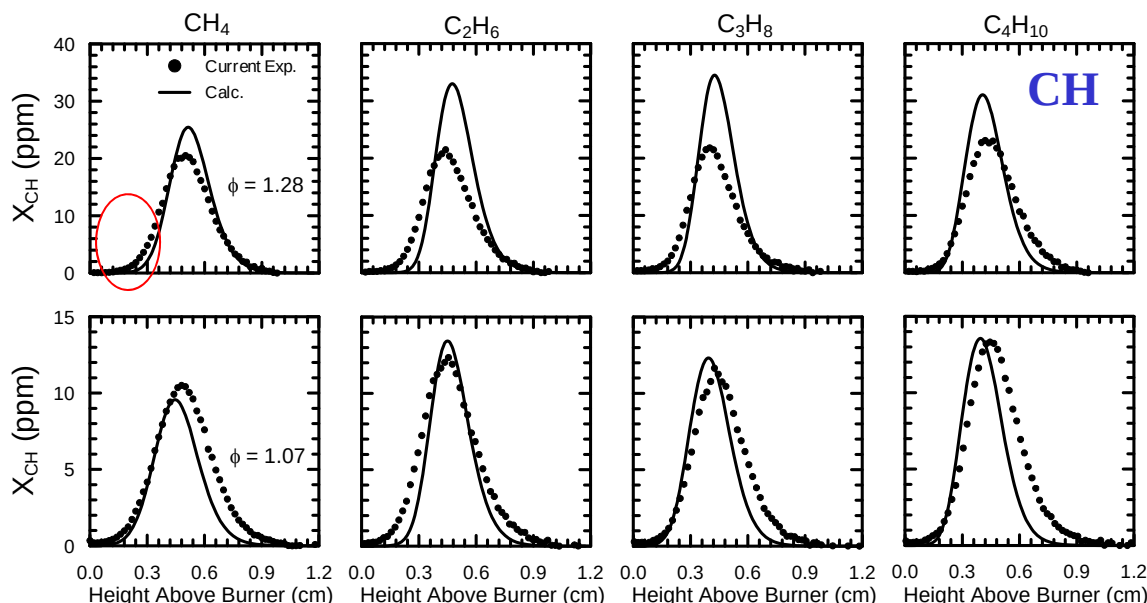
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Spatially-resolved measurements of radicals to understand high-temperature flame chemistry, help kinetic model development

Kinetic modeling: GRI-Mech 3.0

We will look at the region upstream of the flame where coupling between plasma kinetics and flame chemistry is most important





Low-pressure flame / plasma measurements (LIF, CRDS)

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Objective: Examine coupling of plasma and combustion kinetics in a 1-D low-pressure flame. Use spatially-resolved species concentration and temperature measurements by **LIF (OH, H, O, and CH)** and **CRDS (HO₂, HCO, CH₃)** to study the effect of quasi-steady (RF) and repetitively pulsed nsec discharge plasmas on low-temperature chemistry and coupling with the flame zone

Outcome: Kinetic mechanism of low-temperature plasma chemical fuel oxidation and energy release, and its effect on flame speed and burn rate. Specifically, boundary between “low-T” and “high-T” chemistry by measuring HO₂ radical concentration, at the conditions when O₂ is electronically excited



CRDS diagnostics will be used in both “test bed” experiments, (I) high-T, high-P nsec discharge plasma cell, and (II) low-P flame / plasma cell



Thrust 2. Kinetic model development and validation

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Task 8: Development and validation of a predictive kinetic model of non-equilibrium plasma fuel oxidation and ignition, using experimental results of Thrust 1

Goal: Identify key mechanisms, reaction, and rates of plasma chemical fuel oxidation processes for a wide range of fuels, pressures, temperatures, and equivalence ratios. This is absolutely essential to predictive capability of the model.



Current state of the art: hydrocarbon-air, low-temperature plasma chemistry kinetic model

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- Air plasma model: equations for ground state species (N , N_2 , O , O_2 , O_3 , NO , NO_2 , N_2O), charged species (electrons and ions), and excited species ($\text{N}_2(\text{A}^3\Sigma)$, $\text{N}_2(\text{B}^3\Pi)$, $\text{N}_2(\text{C}^3\Pi)$, $\text{N}_2(\text{a}'^1\Sigma)$, $\text{O}_2(\text{a}^1\Delta)$, $\text{O}_2(\text{b}^1\Sigma)$, $\text{O}_2(\text{c}^1\Sigma)$, $\text{N}(\text{D})$, $\text{N}(\text{P})$, $\text{O}(\text{D})$) produced in the plasma.
- Two-term expansion Boltzmann equation for plasma electrons
- Fuel-air plasma: model combined with GRI Mech 3.0 C_xH_y oxidation mechanisms, supplemented with fuel dissociation by electron impact and in reactions with electronically excited nitrogen
- Peak E/N adjusted for pulse energy to be same as predicted by the nanosecond pulse discharge model

We have absolutely no reason to trust the model predictions: GRI Mech 3.0 (or any other combustion mechanism) is not designed to work at low temperatures (starting at $T=300\text{ K}$)

Confidence in the model can be provided only by detailed kinetic measurements such as discussed in Thrust 1 plan

Here is what we know so far: dominant radical and energy release processes in C₂H₄-air predicted by the model

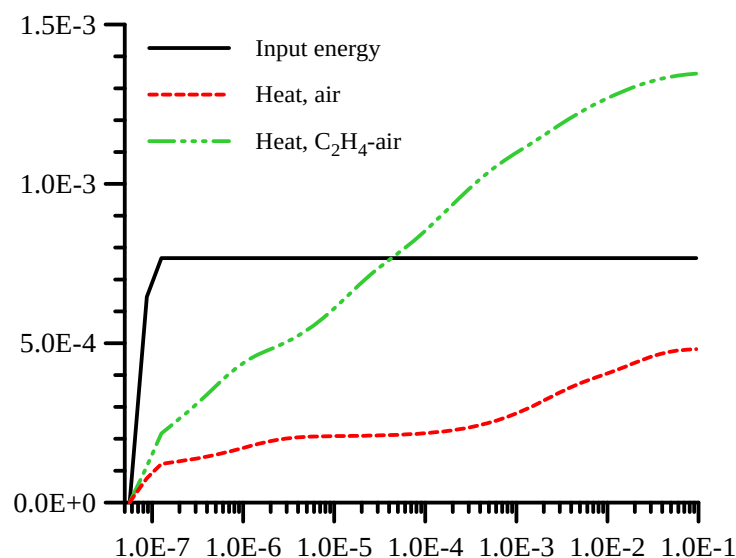
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O atom generation		
N ₂	+ e ⁻	= N ₂ (A ³ Σ) + e ⁻
N ₂	+ e ⁻	= N ₂ (B ³ Π) + e ⁻
N ₂	+ e ⁻	= N ₂ (C ³ Π) + e ⁻
N ₂	+ e ⁻	= N ₂ (a ¹ Σ) + e ⁻
O ₂	+ e ⁻	= O(³ P) + O(³ P, ¹ D) + e ⁻
N ₂ (C ³ Π)	+ O ₂	= N ₂ (a ¹ Σ) + O ₂
N ₂ (a ¹ Σ)	+ O ₂	= N ₂ (B ³ Π) + O ₂
N ₂ (B ³ Π)	+ O ₂	= N ₂ (A ³ Σ) + O ₂
N ₂ (A ³ Σ)	+ O ₂	= N ₂ + O + O
Fuel dissociation		
C ₂ H ₄	+ e ⁻	= products + e ⁻
N ₂ (A ³ Σ)	+ C ₂ H ₄	= N ₂ + C ₂ H ₃ + H
N ₂ (B ³ Π)	+ C ₂ H ₄	= N ₂ + C ₂ H ₃ + H
N ₂ (C ³ Π)	+ C ₂ H ₄	= N ₂ + C ₂ H ₃ + H
N ₂ (a ¹ Σ)	+ C ₂ H ₄	= N ₂ + C ₂ H ₃ + H
O atom decay		
O	+ C ₂ H ₄	= CH ₃ + HCO
O	+ C ₂ H ₄	= H + CH ₂ CHO
C ₂ H ₃	+ O ₂	= HCO + CH ₂ O
C ₂ H ₃	+ O ₂	= O + CH ₂ CHO
O	+ O ₂ + M	= O ₃ + M
O	+ O ₃	= O ₂ + O ₂

Fuel energy release		
O	+ CH ₂ CHO	= H + CH ₂ + CO ₂
H	+ O ₂ + M	= HO ₂ + M
O	+ HO ₂	= OH + O ₂
OH	+ HO ₂	= O ₂ + H ₂ O
OH	+ C ₂ H ₄	= C ₂ H ₃ + H ₂ O
HO ₂	+ CH ₃	= OH + CH ₃ O
CH ₃ O	+ O ₂	= HO ₂ + CH ₂ O
O ₂	+ CH ₂ CHO	= OH + HCO + HCO
HCO	+ O ₂	= HO ₂ + CO
HO ₂	+ HO ₂	= O ₂ + H ₂ O ₂
CH ₂	+ O ₂	= H + H + CO ₂

Pulse energy balance, J



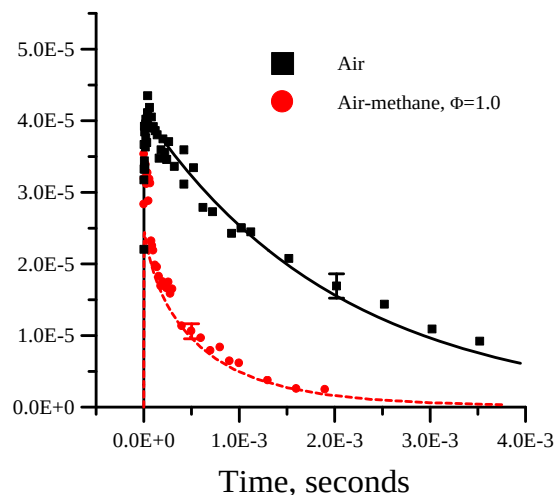
Time, seconds

Model validation summary: so far so good... ... but no surprise if the model fails at some point

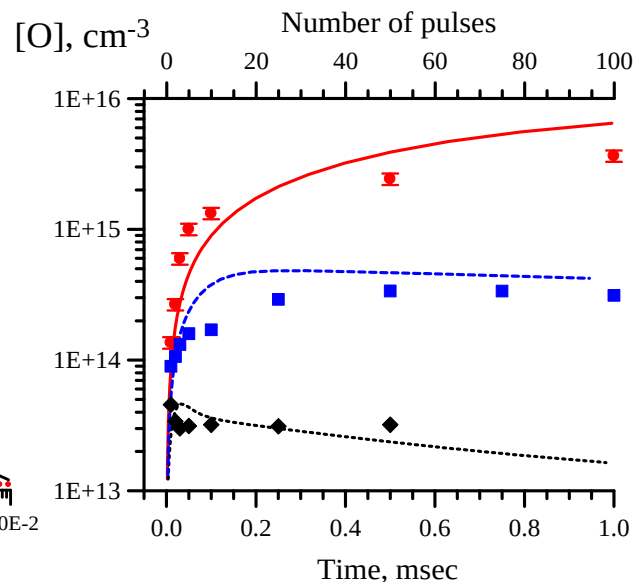
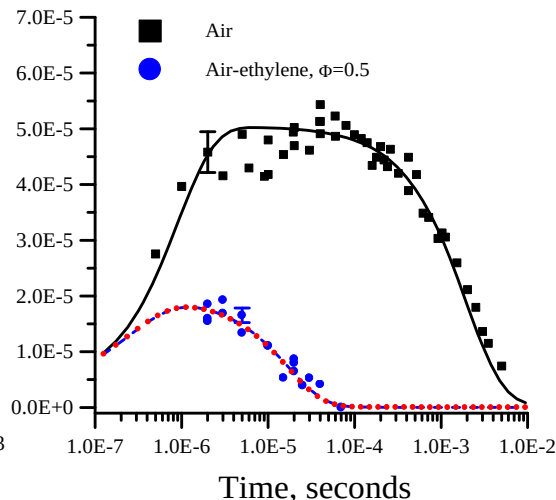
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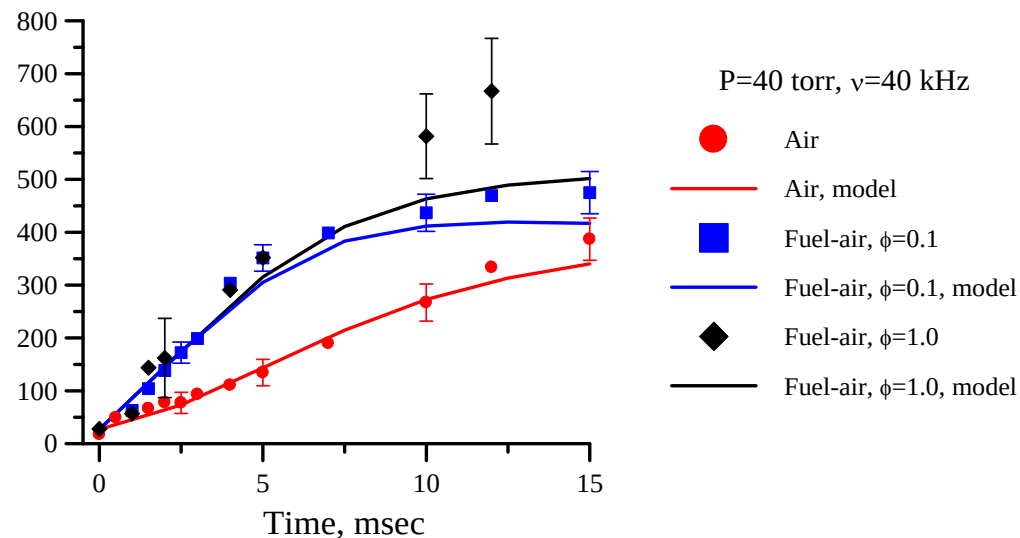
O atom mole fraction



O atom mole fraction



T, °C



Need a lot more data from Thrust 1 for extensive model validation

Outcome: a self-consistent low-temperature fuel-air plasma chemical mechanism



Thrust 3. Experimental and modeling studies of fundamental nonequilibrium discharge processes

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Task 10: Characterization and modeling of nsec pulse discharges

Goal: Prediction of E/N and electron density in the plasma, individual pulse energy coupled to the plasma, and their scaling with pressure, temperature, pulse waveform, and mixture composition



Two-pronged approach to plasma assisted ignition modeling

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Predictive modeling of energy release rate and ignition delay time in low-temperature, repetitive nanosecond pulse fuel-air plasmas requires:

- **E/N in the plasma, individual pulse energy coupled to the plasma, and their scaling with pressure, temperature, pulse waveform, and mixture composition**
- **Air plasma and fuel-air plasma chemistry: reactions among ground state species, excited species and radicals generated in the plasma, and their effect on energy release rate**

These two problems require separate analysis:

- **Nsec pulse plasma / sheath models cannot incorporate detailed reactive plasma chemistry: too many species (~100) and reactions (~1,000)**
- **Detailed plasma chemistry models (quasi-neutral) cannot incorporate repetitive, nsec time scale sheath dynamics and plasma shielding**

Approach:

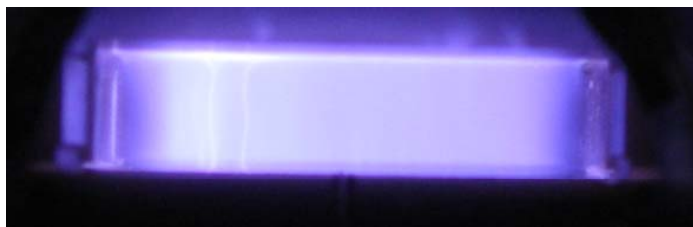
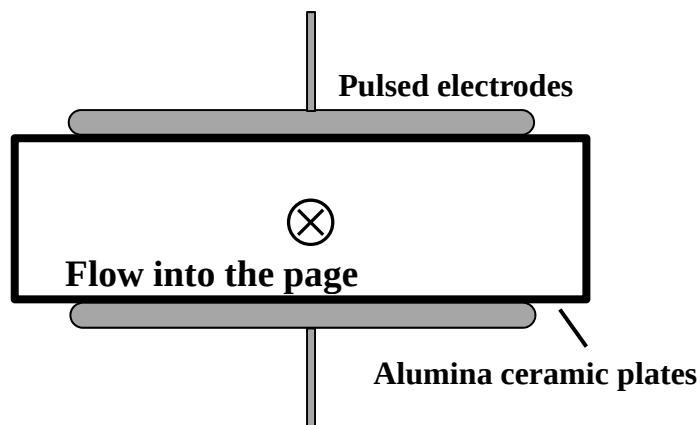
- **Predict plasma E/N and coupled pulse energy using nsec pulse plasma / sheath model**
- **Incorporate results into fuel-air plasma chemistry model**

Previous results:

Repetitive nsec discharge pulse energy measurements

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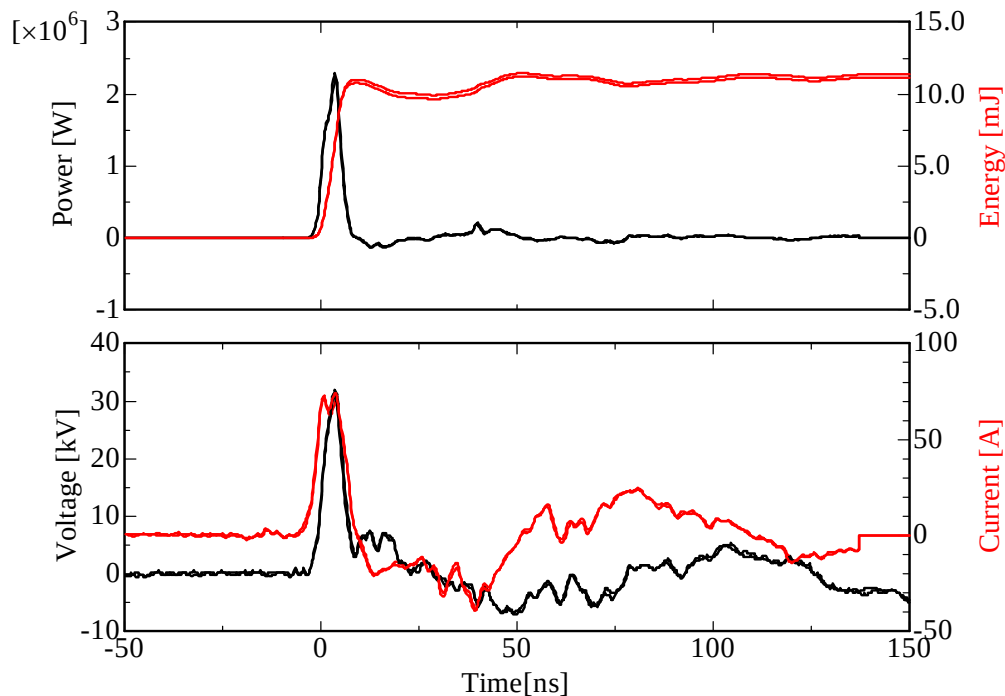
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Nitrogen, **P=300 torr**, $\nu=100$ kHz



Nitrogen, **P=650 torr**, $\nu=100$ kHz



Nitrogen, **P=350 torr**, $\nu=100$ kHz
0.3 seconds after start (pulse # 30,000)

Pulse energy 11 mJ/pulse
Discharge power 110 W

What are the electric field and the electron density?

Previous results:

Analytic nsec pulse discharge plasma / sheath model

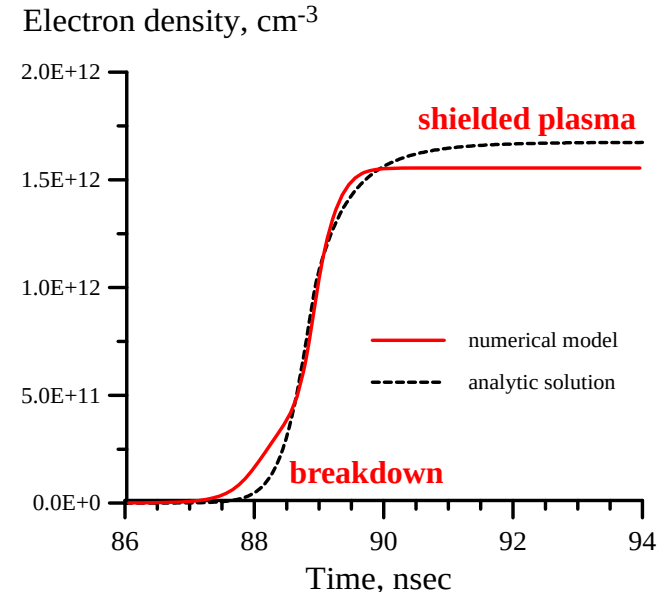
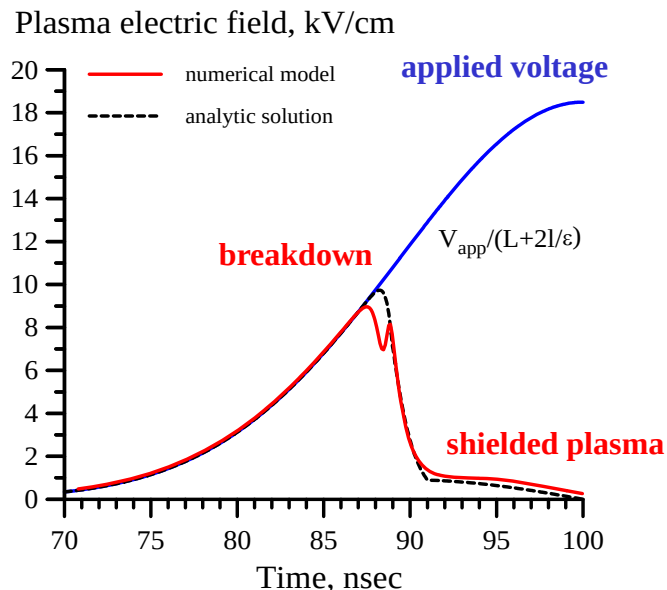
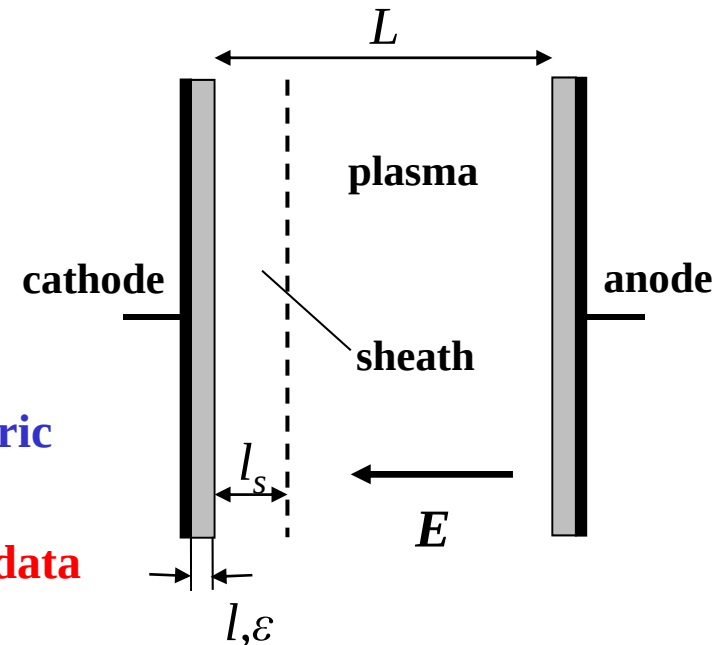
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- Equations for electron and ion number density
- Poisson equation for the electric field
- Plane-to-plane discharge geometry
- Voltage pulse: Gaussian fit to experimental waveform
- Dielectric plate charging / plasma shielding

Analytic solution: time-dependent electron density and electric field in the plasma, coupled pulse energy

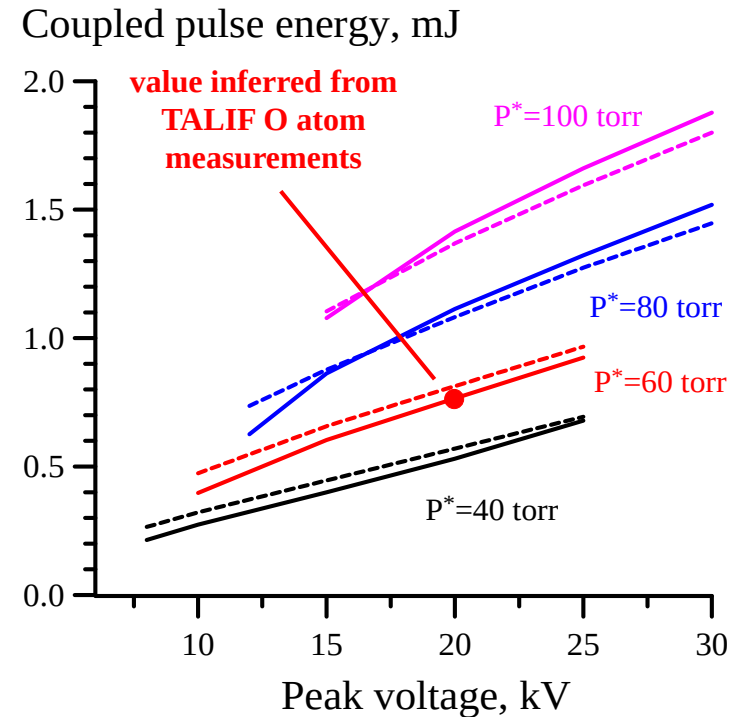
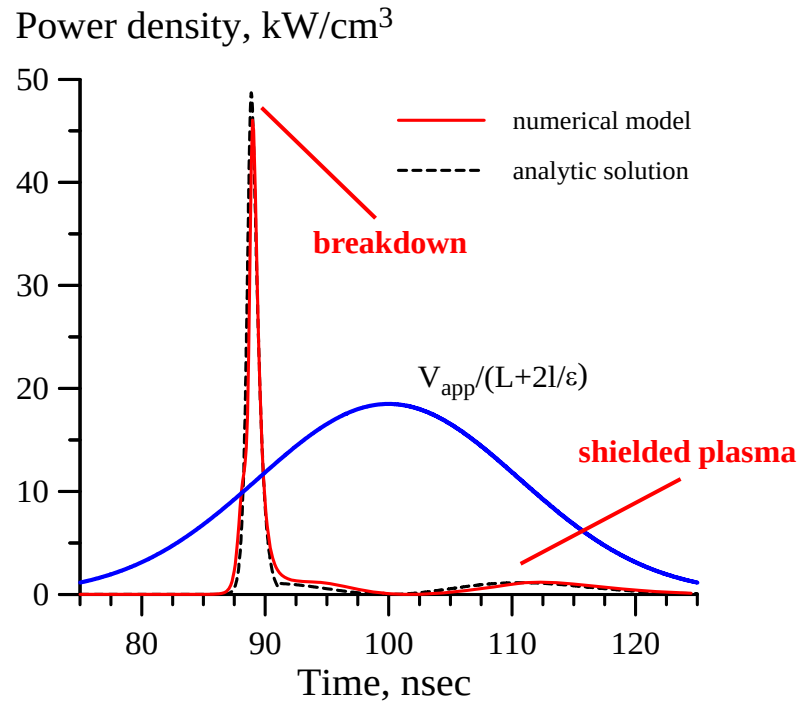
Excellent agreement with numerical solution, experimental data



Previous results: Analytic nsec pulse discharge plasma / sheath model

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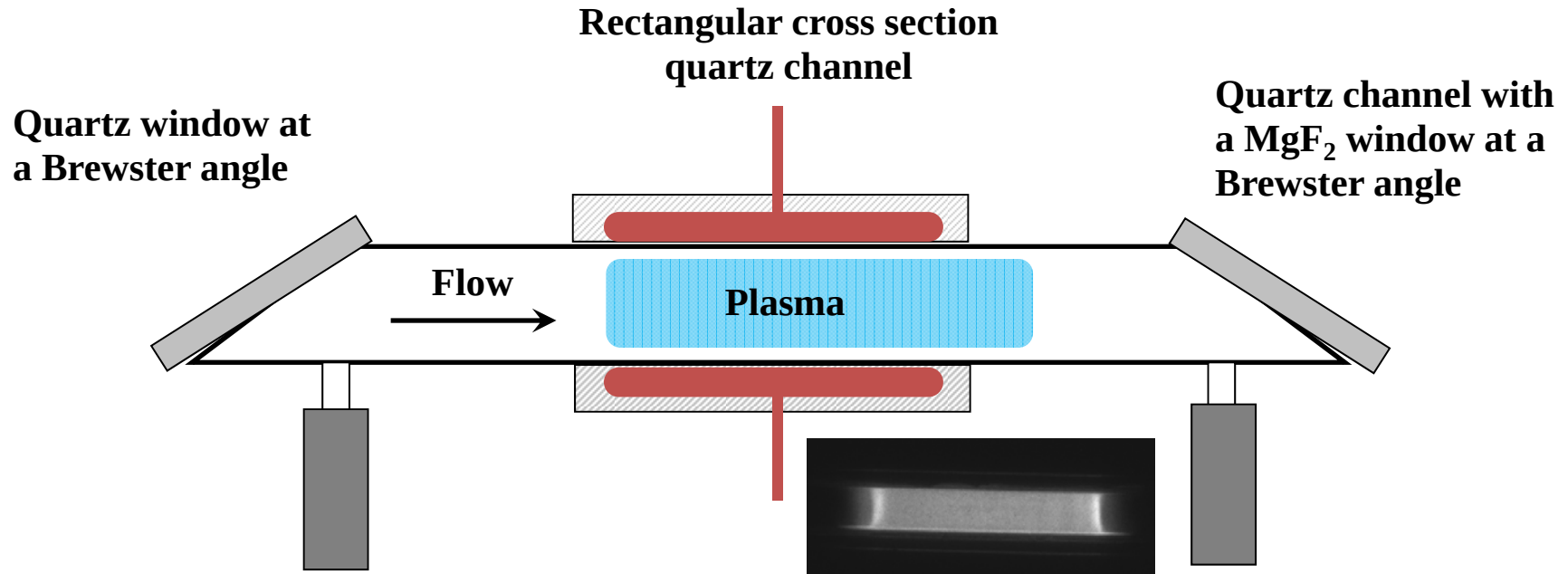
$$Q_{total} = Q_{break} + Q_{after} \approx \frac{1}{2} C_{load} V_{peak}^2 \left[\left(\frac{V_0}{V_{peak}} \right)^2 + \frac{\sqrt{2\pi}}{v_{RC} \tau_{pulse}} \right]$$

- Coupled pulse energy scales with the number density, can be increased by increasing peak voltage, reducing pulse duration
- Excellent agreement with numerical solution, experimental data

Electric field and electron density measurements: CARS, Thomson scattering

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Entire test section mounted on a translation stage for spatially resolved measurements.

Objective: measure time- and space-resolved electric field and electron density in nsec pulse discharge plasmas using psec CARS and Thomson scattering; comparison with the model

Outcome: predictive capability for electron impact kinetic processes in the plasma



Thrust 4. Studies of diffusion and transport of active species in representative 2-D reacting flow geometries

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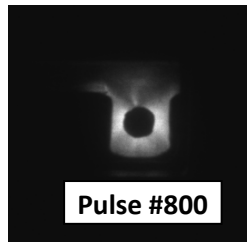
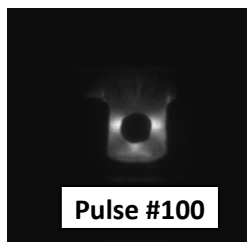
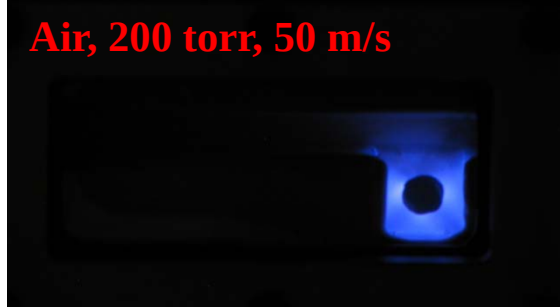
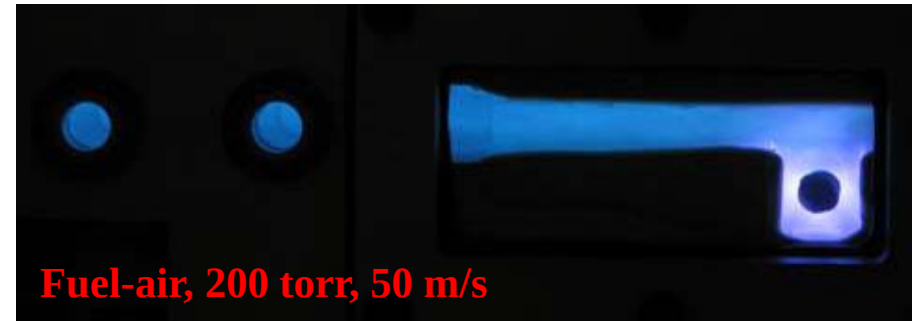
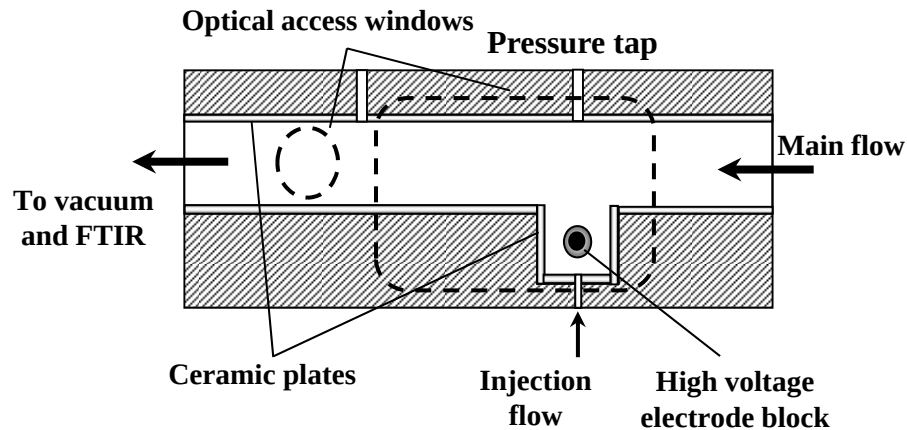
Task 12: Ignition and flameholding in nonequilibrium plasma cavity flows at low static temperatures

Goal: Determine viable approaches to flameholding in high-speed flows using low-temperature plasmas. We simply cannot process the entire flow with the plasma!

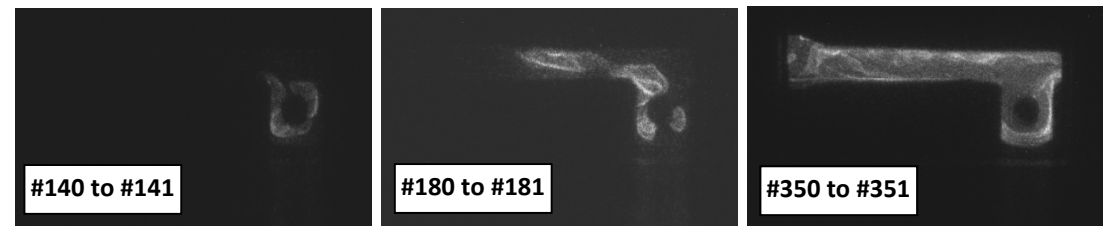
Previous results: cavity ignition in premixed ethylene-air flows by nsec plasma (25 kV, 20 nsec, 40 kHz)

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Air, 150 torr, 25 m/s



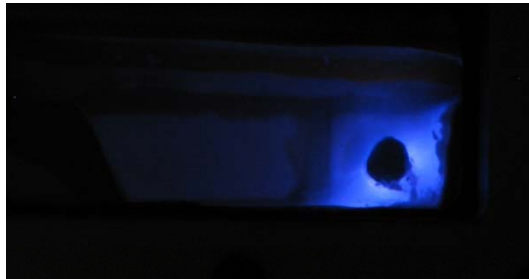
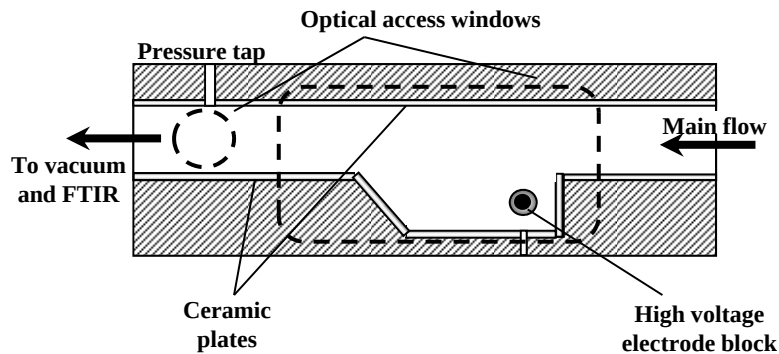
Fuel-air, 150 torr, 25 m/s

Diffuse plasma in air, filamentation in fuel-air during ignition, diffuse plasma after ignition

Previous results: cavity ignition and flameholding in premixed and non-premixed ethylene-air flows by nsec plasma

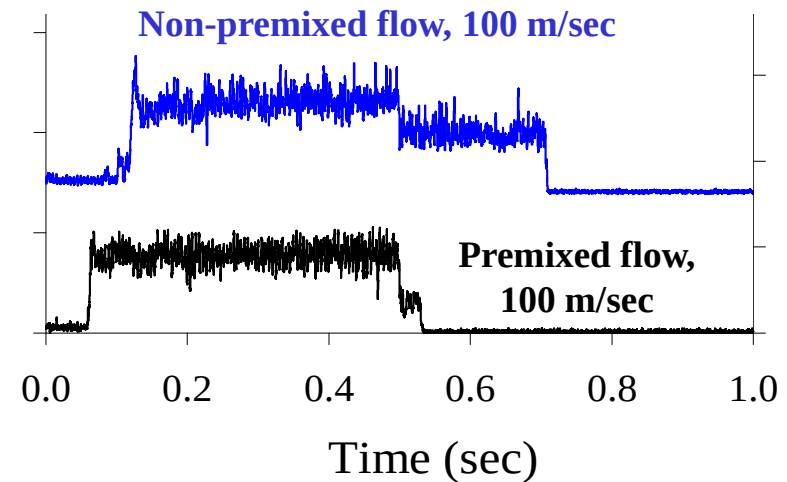
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Fuel-air, 175 torr, 85 m/s

Intensity (arbitrary units) **OH emission**



- Ignition and stable flameholding in both premixed and non-premixed flows up to 100 m/sec (global $\phi=1$ in both cases)
- 80-90% burned fuel fraction
- Plasma power ~ 100 W, combustion energy release 35 kW
- After ignition, plasma needs to be “on” at all times (flame blow-off without plasma)



Ignition and flameholding in nonequilibrium plasma cavity flows

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Objectives:

- Further studies of cavity ignition and flameholding by repetitive nsec pulse plasmas in fuel injection flows (hydrogen and hydrocarbons)
- High frame rate (10-20 kHz) NO and OH PLIF imaging of ignition process using burst mode laser
- Increasing flow velocity beyond 100 m/sec, operating at low global equivalence ratios ($\phi=0.1-0.2$)
- Comparison with kinetic modeling calculations using reduced plasma chemical ignition mechanism. Plasma flameholding mechanism after ignition – thermal or not?

Outcome: Demonstration of true predictive capability of the model