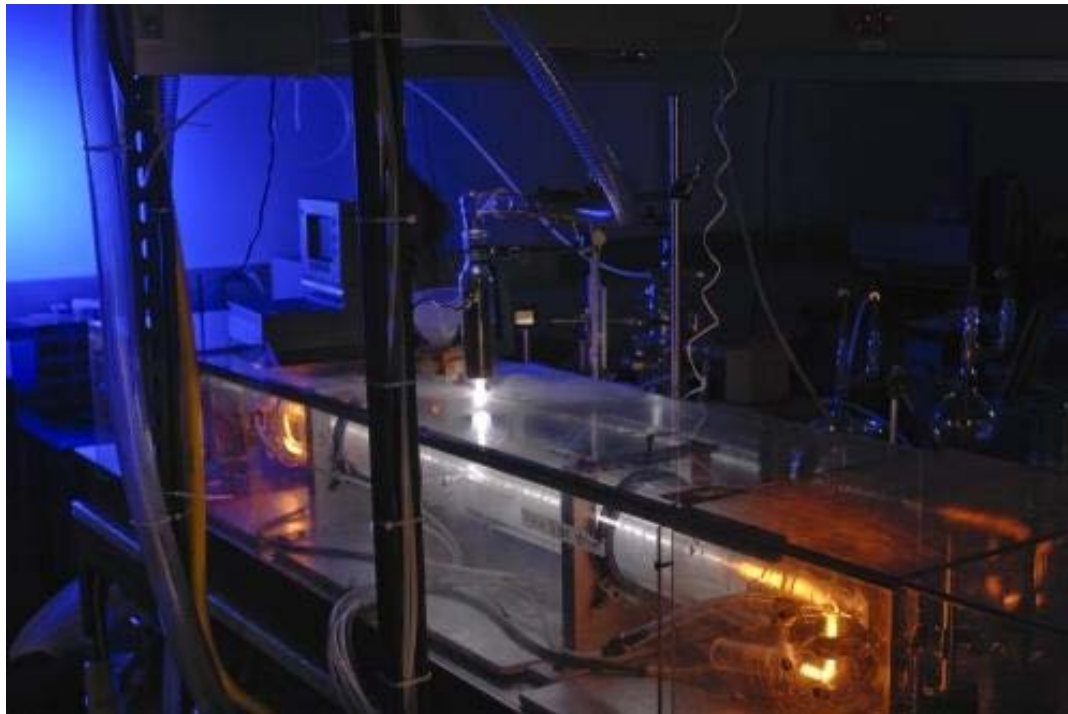


Laser diagnostics for electric field measurements in air plasmas, plasma-enhanced flames, and atmospheric pressure plasma jets

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*Nonequilibrium Thermodynamics Laboratory
Department of Mechanical and Aerospace Engineering*



Electric Field in Plasmas: Why Do We Care?

- **Electric field controls energy partition in the plasma (vibrational and electronic excitation, molecular dissociation), reactive species generation, temperature rise**
- **Excited metastable species and reactive radicals: strong effect on plasma-induced chemistry, emission from nonequilibrium high-speed flows**
- **Focus on ns pulse discharges: stable at high pressures, efficient generation of excited species and reactive radicals**
- **Need for non-intrusive, spatially resolved, time-resolved measurements of electric field and species densities in transient plasmas**
- **Insight into kinetics of ionization, charge transport, plasma chemical syntheses (such as fuel reforming and plasma catalysis), effect on the flow field**

Energy Partition in Air Plasma vs. Electric Field

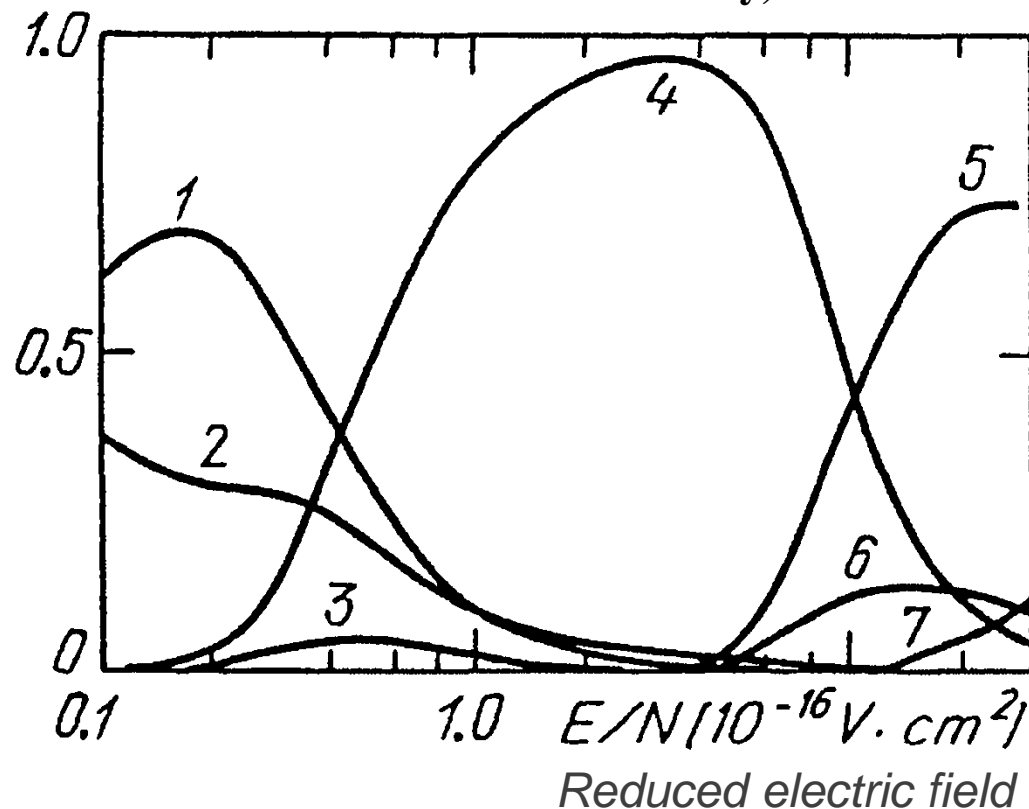
Yu. Raizer, Gas Discharge Physics, Springer, 1991

Quasi-steady-state discharges (DC, RF, MW): low E/N

(4) N₂ vibrational excitation:

Low reactivity, slow thermalization

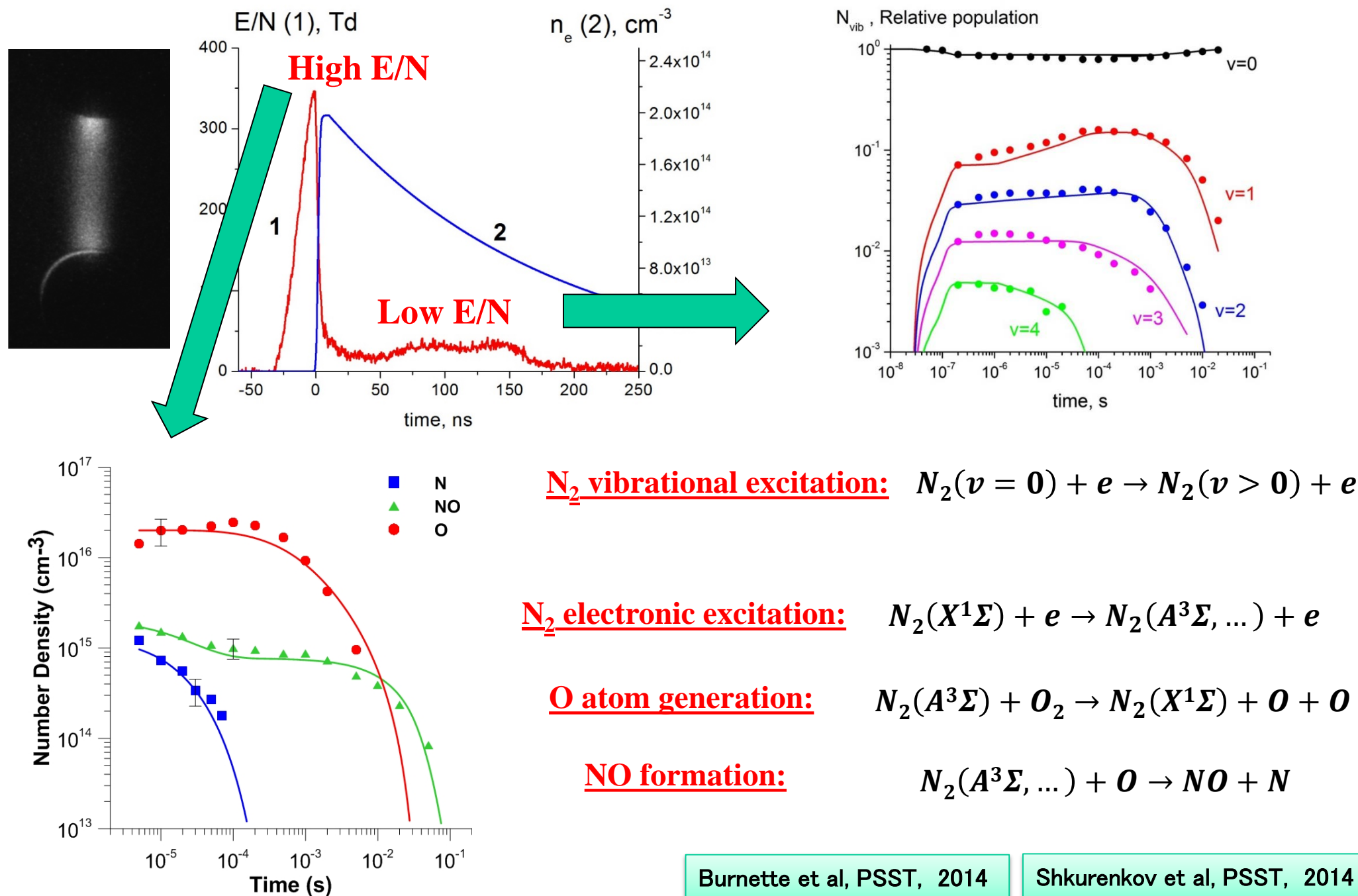
Energy fraction



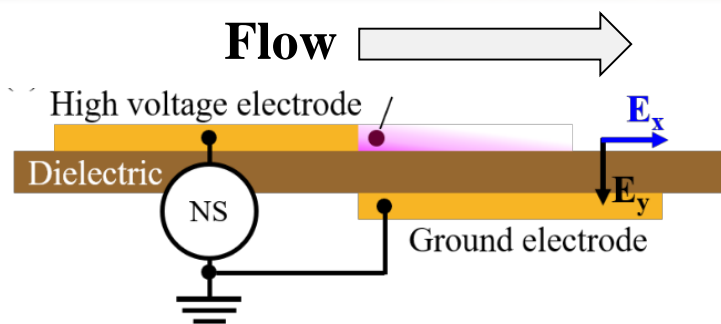
Pulsed discharges (NS, AC DBD,): high E/N
(5,6) N₂, O₂ electronic excitation, dissociation,
High reactivity, rapid thermalization

- Reduced electric field, E/N , controls input energy partition in the discharge
- Rates of electron impact processes: depend on T_e , E/N exponentially

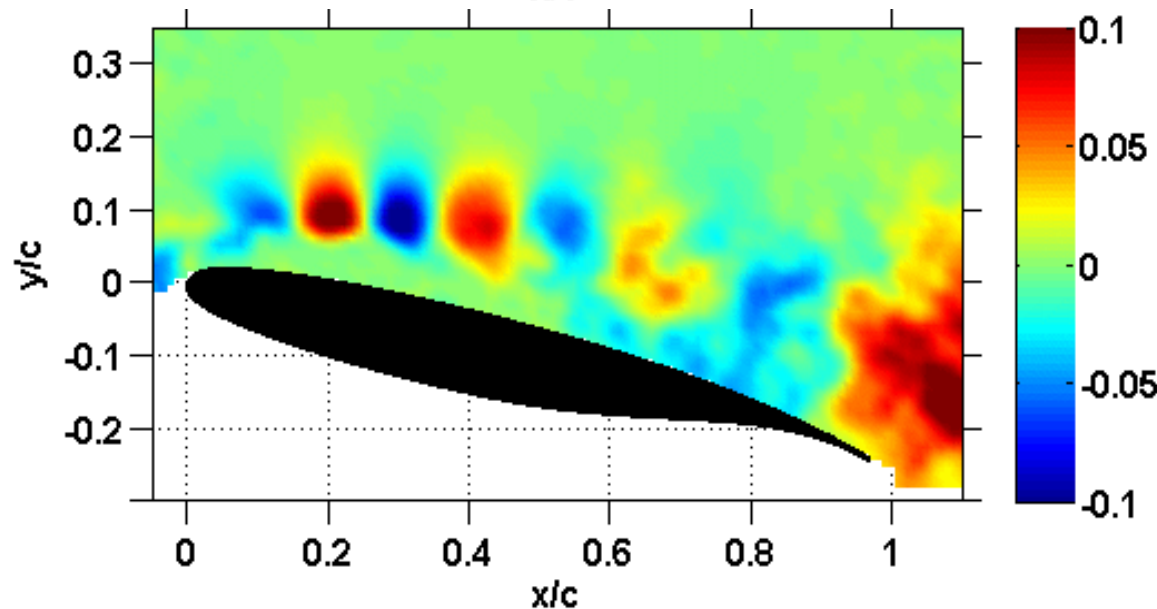
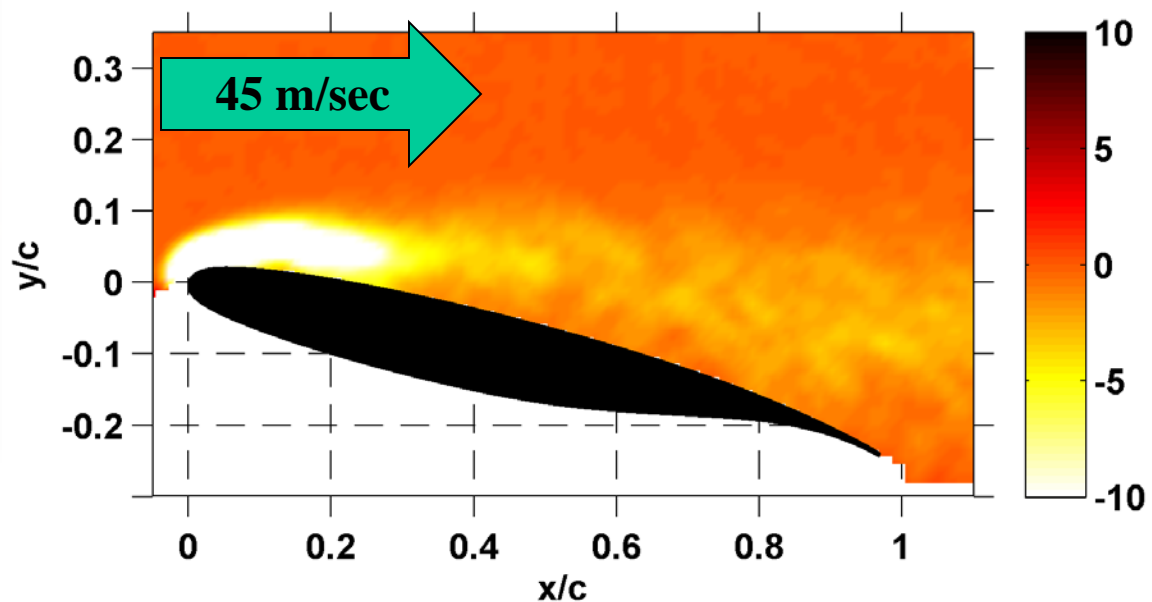
Air Plasma Chemistry: Both Low and High E/N Pathways Contribute



Ns Pulse Surface Plasma Actuators: Efficient High-Speed Flow Control



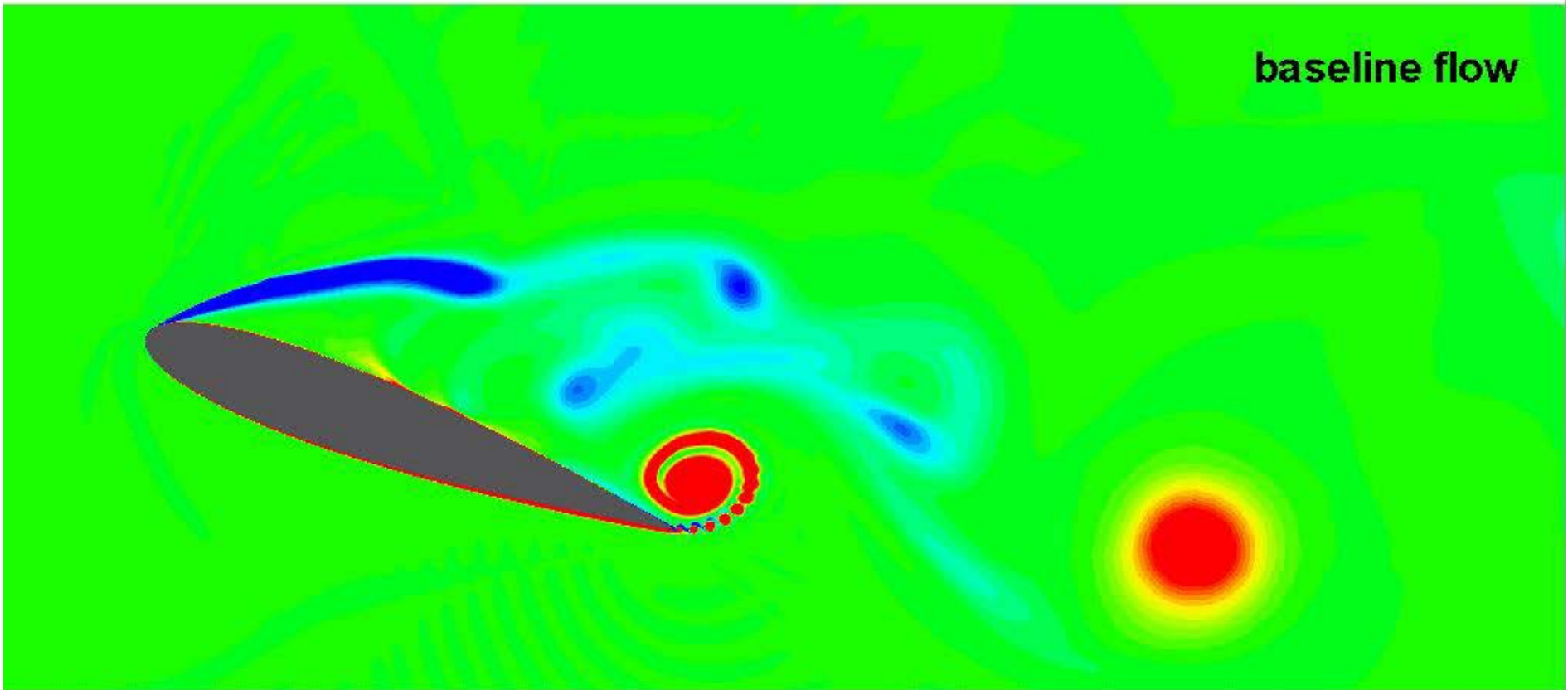
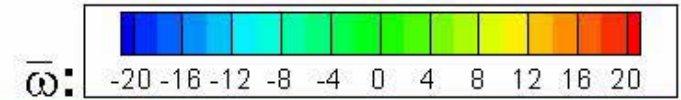
- Every ns discharge pulse produces a spanwise “vortex tube”
- Enhanced mixing with free stream boundary layer reattachment
- Effect detected up to $u=96$ m/s



Plasma / CFD Modeling Predictions

Baseline and forced flows, $Re = 1.2 \cdot 10^6$

$Re = 1.2 \times 10^6$: $U_\infty = 93 \text{ m/s}$, $c = 20.32 \text{ cm}$, $\alpha = 20^\circ$



Vortex formation controlled by rapid heating in the plasma (high E/N process)

Need to know time-resolved electric field to predict this accurately

E-FISH: How We Measure Electric Field



- Well-known technique, developed in 1970s
- Applied to plasma diagnostics only recently (since 2018)

Electric Field Induced Second Harmonic (E-FISH) Generation

- Only one pulsed laser is needed (Nd:YAG, Ti:Sapphire, ...)
- Not species-specific, can be used in any high-pressure plasma
- Signal is in visible/UV: straightforward detection
- Signal is generated as a coherent beam
- Signal proportional to electric field squared, laser intensity squared: ps and fs lasers are the most effective
- Signal polarization same as field direction: E_x , E_y are measured separately
- Taking data requires days, not years

E-FISH: How Does It Work?

- External electric field, E^{ext} , induces a dipole moment in molecules or atoms
- Laser field generates oscillating polarization, with driving force $\sim [E^{(\omega)}]^2$
- Coherently oscillating dipoles launch a coherent wave at 2ω

$$I_i^{(2\omega)} \sim [\chi_{ijkl}^{(3)}(2\omega, 0, \omega, \omega) E_j^{\text{ext}} E_k^{(\omega)} E_l^{(\omega)}]^2 L^2 \left[\frac{\sin(\Delta k \cdot L/2)}{\Delta k \cdot L/2} \right]^2$$

$I^{(2\omega)}$ – SH signal; $\chi^{(3)} \sim N$ – nonlinear susceptibility;

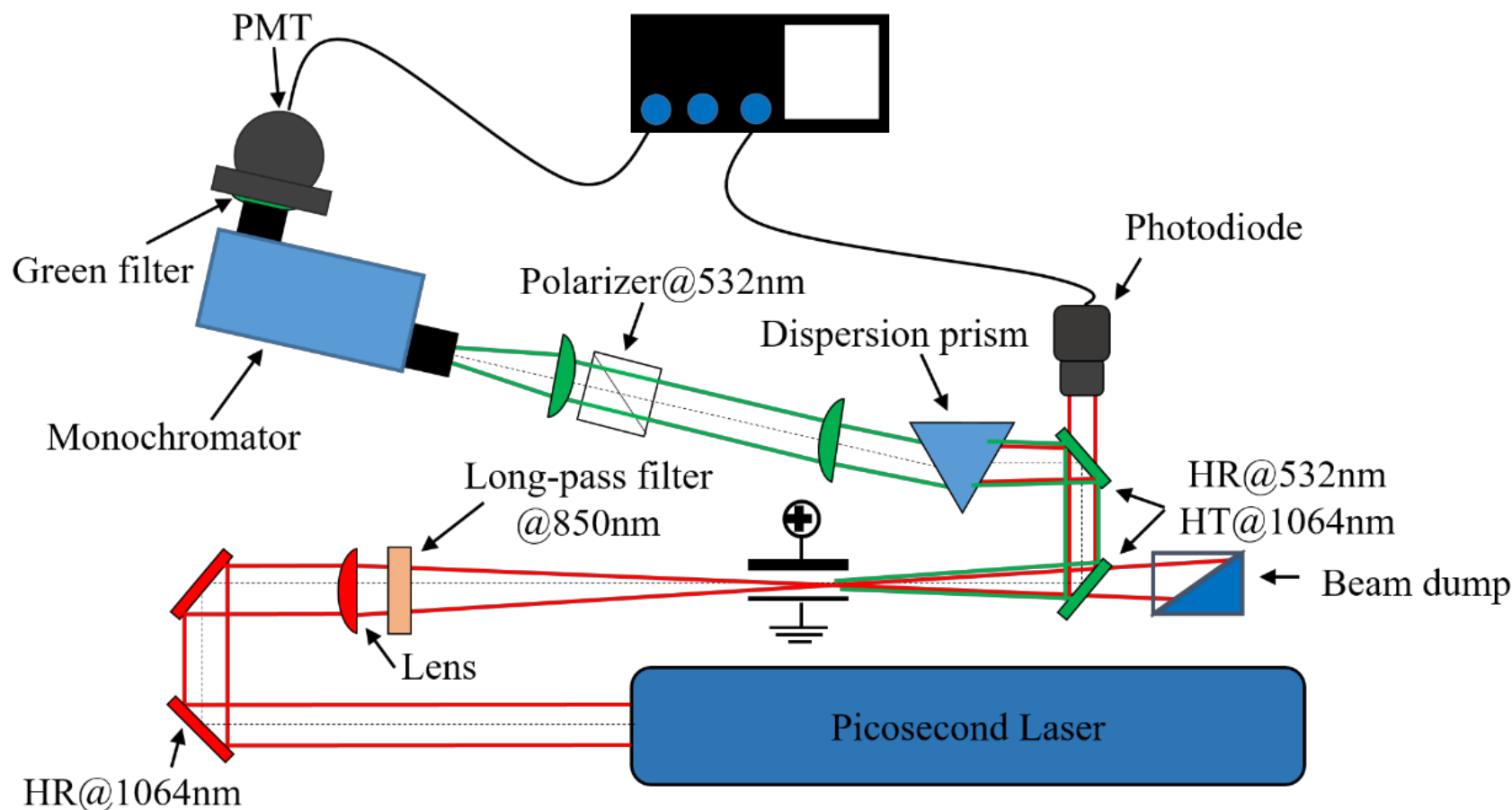
L – interaction length (controlled by confocal parameter of the lens)

Phase matching parameter Δk limits coherent growth of the signal

$$\Delta k = k(\omega) - k(2\omega) = \frac{2\omega \cdot n(2\omega) - \omega \cdot n(\omega)}{c}$$

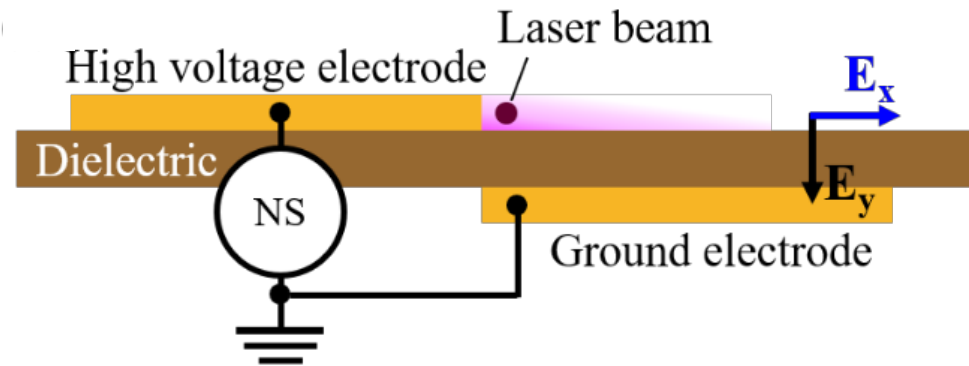
Coherence length in ambient air, for 1064 nm pump beam: $L_c = \pi/\Delta k = 6 \text{ cm}$

E-FISH Experiment Schematic

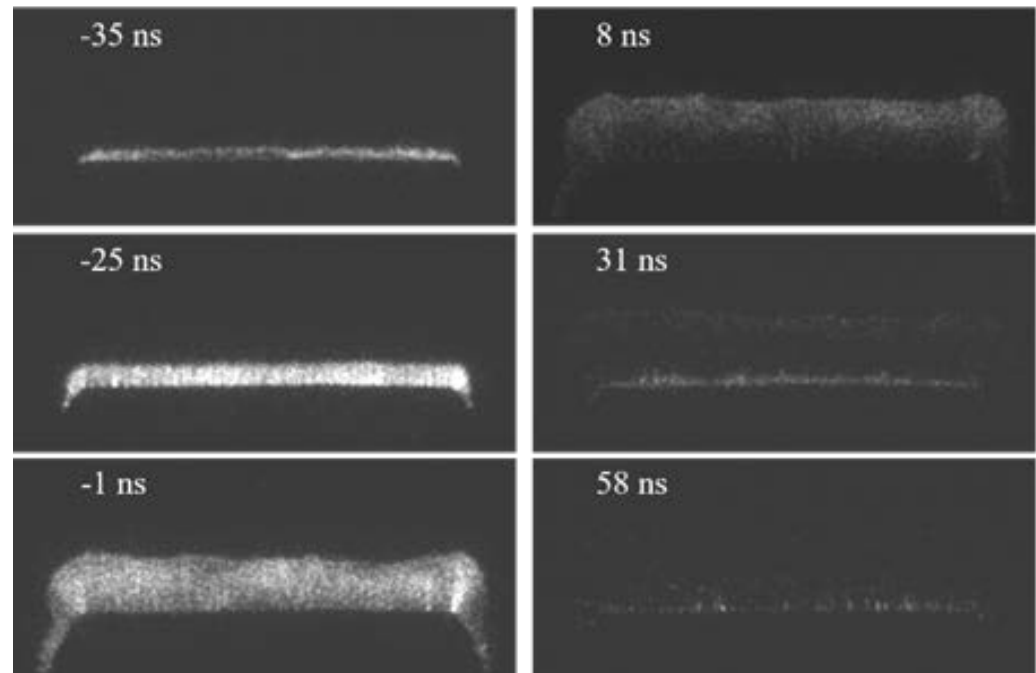


- 30 ps laser pulse, 2-10 mJ/pulse at 1064 nm, generating second harmonic at 532 nm
- Second harmonic generation vs. compared to CARS-like 4-wave mixing:
10 times more signal at 10 times less laser power

Ns Pulse Surface DBD Plasma Actuator



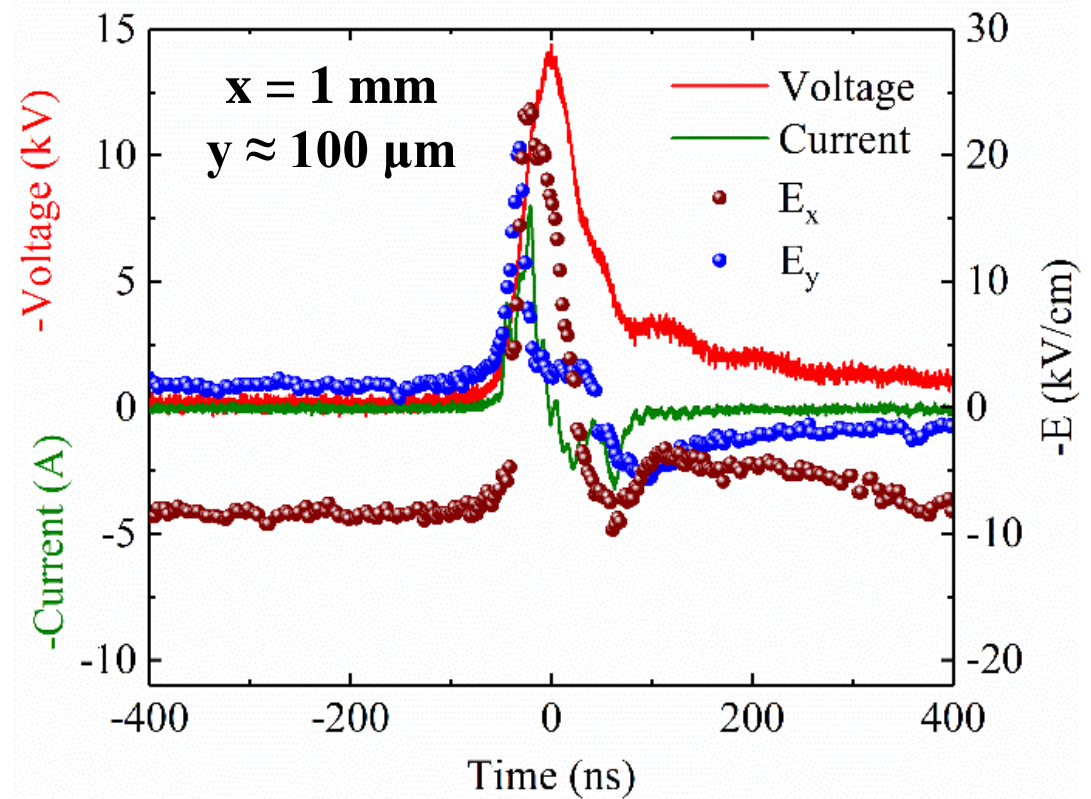
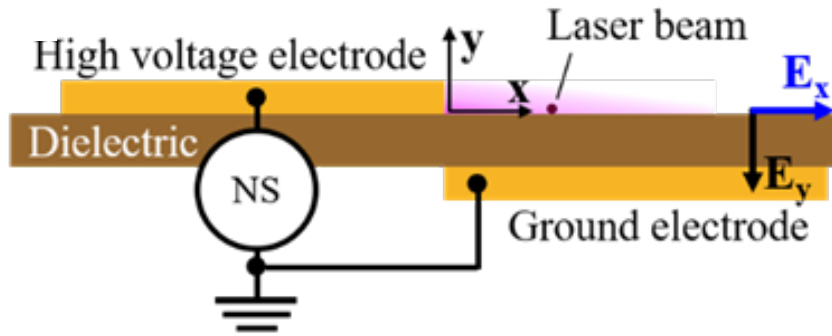
Side view



Top view

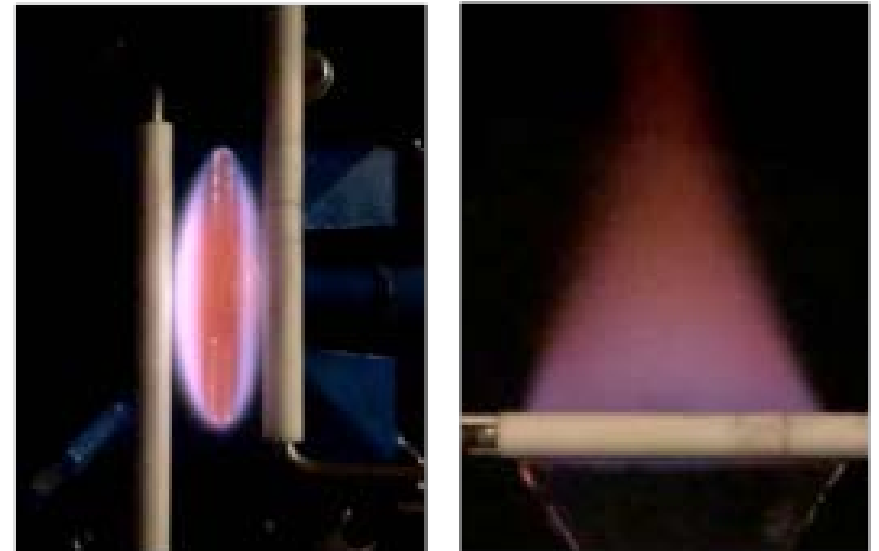
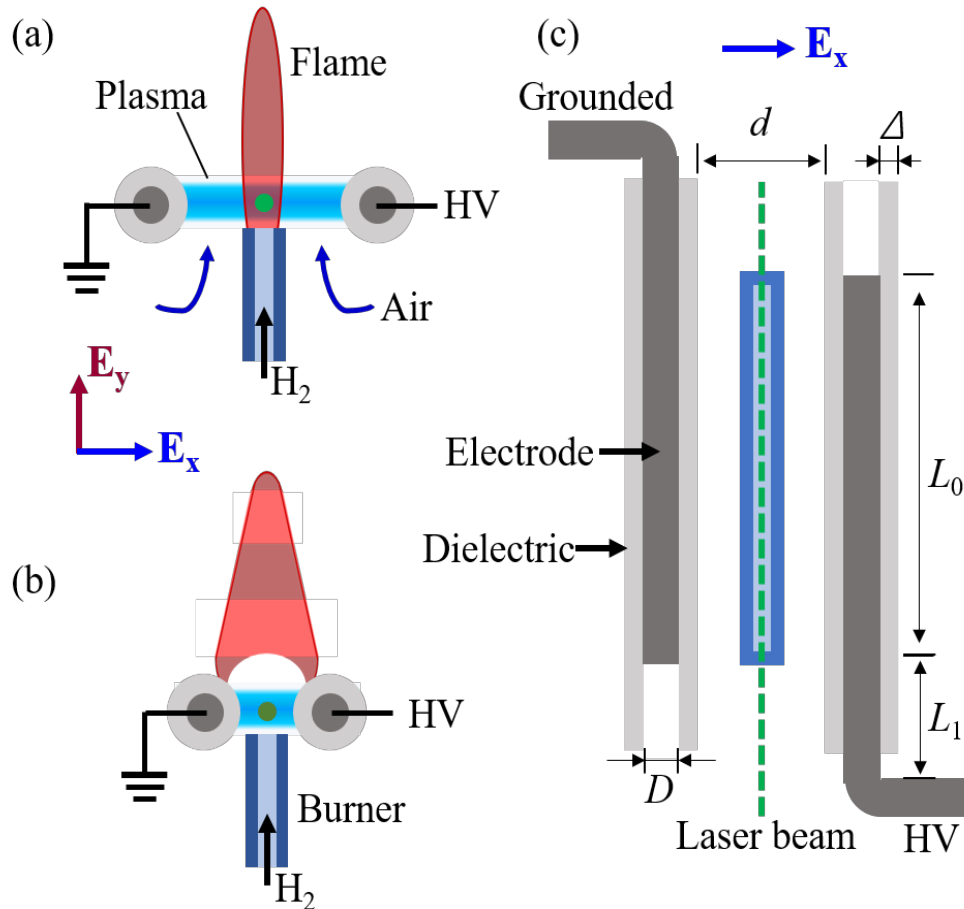
- Room air, dielectric plate 0.6 mm thick, rep rate 20 Hz, peak voltage 13 kV
- Negative polarity, diffuse surface ionization wave

Results: Ns Pulse Surface DBD Plasma Actuator



- Field offset due to residual charge accumulation from previous pulse
- Field rise with applied voltage, reduction after breakdown (plasma self-shielding)
- Field reversal during voltage reduction (dielectric surface charging)
- $E(t)/N$ controls temperature rise in the plasma layer, vortex formation in the flow

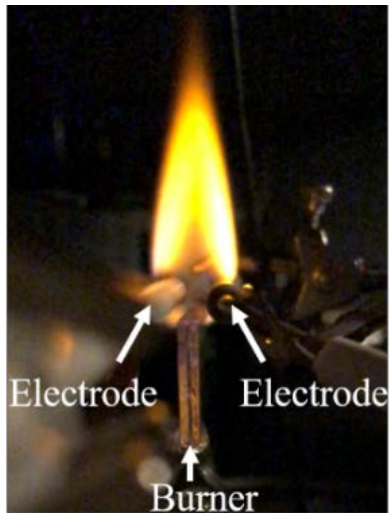
Two-Dimensional H₂ Diffusion Flame



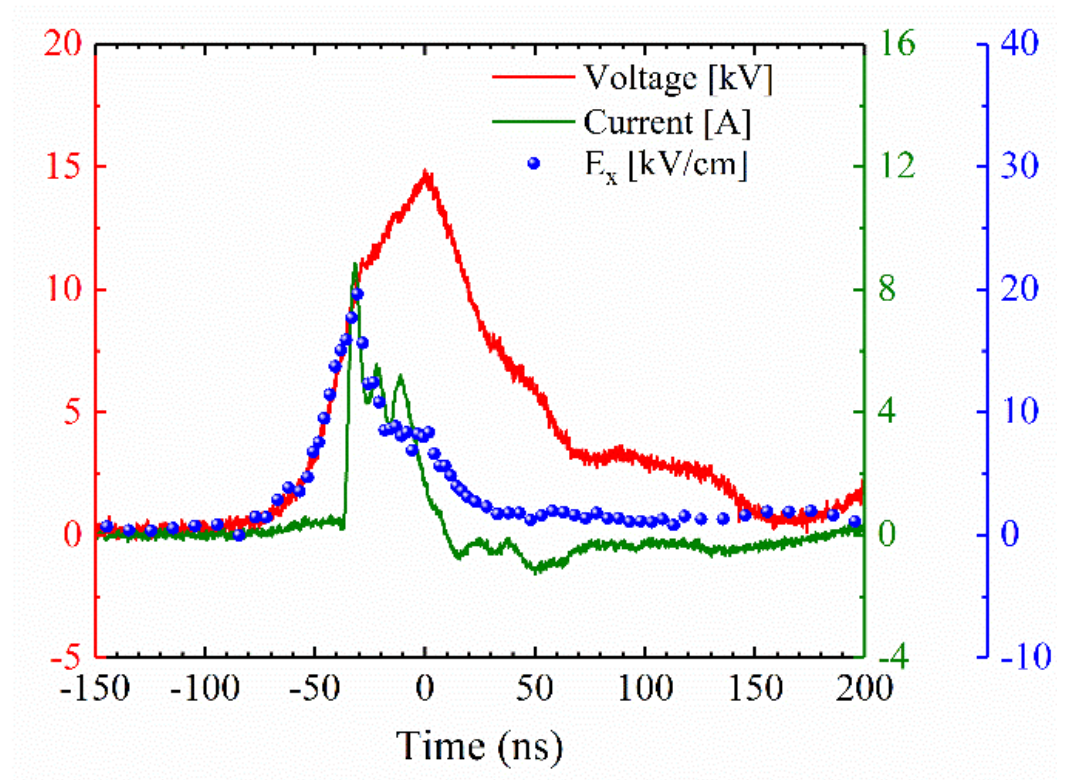
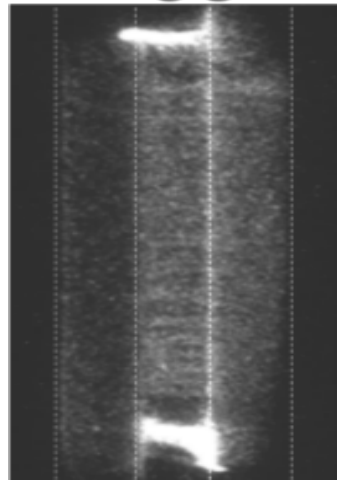
No plasma

- Quasi-two-dimensional diffusion flame
- Electrodes in ceramic tubes, powered by AC and ns pulse voltage waveforms
- Flame can be attached to burner or electrode sleeves

Results: Ns Pulse Discharge in H₂ Diffusion Flame

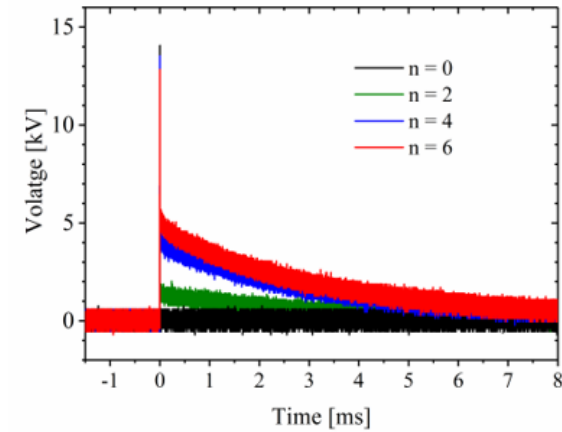
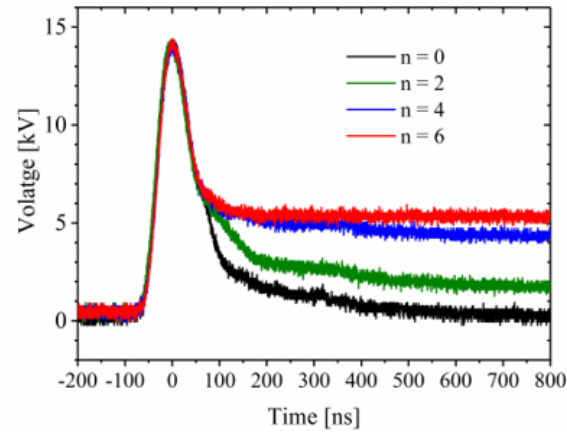
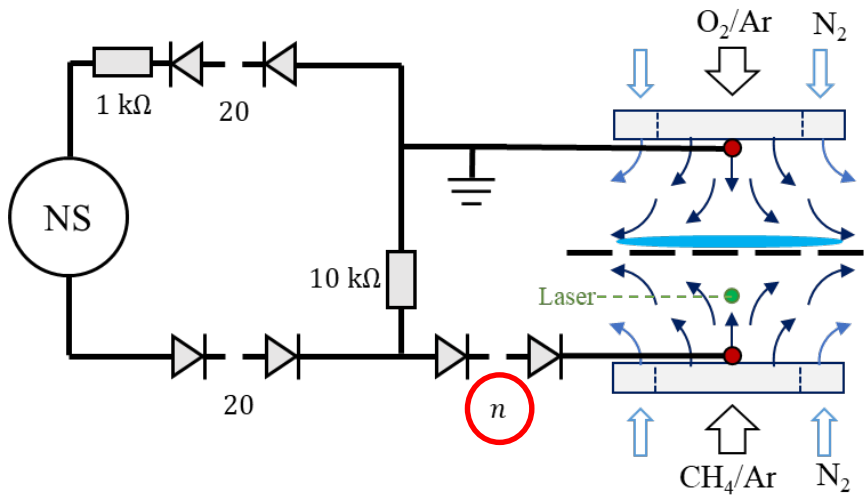


Top view
400 ns gate



- Flame attached to top of electrode tubes, **field measured in H₂ plasma, T=370 K**
- No field offset: method is self-calibrating, no need to know mixture composition
- Energy coupled at $E=9-19$ kV/cm, $E/N = 50-100$ Td: efficient H₂ dissociation by electron impact, generation of H atoms
- **Knowledge of $E/N(t)$ is critical for prediction of radical species generation**
- No effect of ns pulse discharge on the flame ☹️

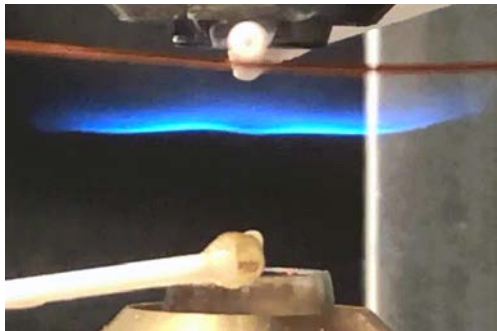
More Interesting Case: Counterflow Flame



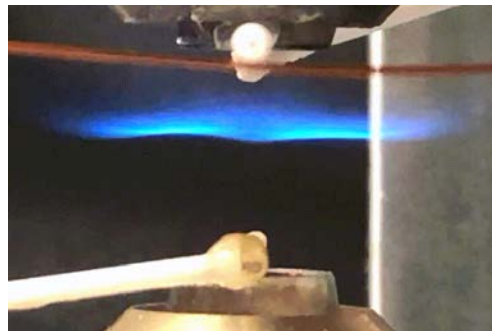
Counterflow, atmospheric pressure $\text{CH}_4\text{-O}_2\text{-Ar}$ flame, N_2 co-flow

Two parallel cylinder electrodes in alumina ceramic sleeves

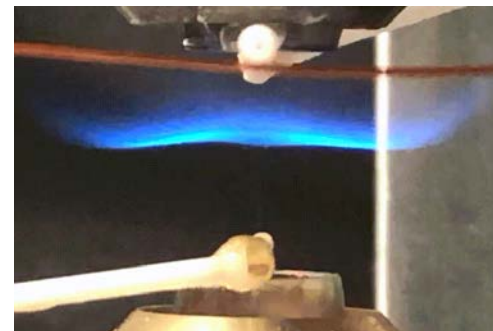
Flame forced by discharge pulse waveforms at 10 Hz



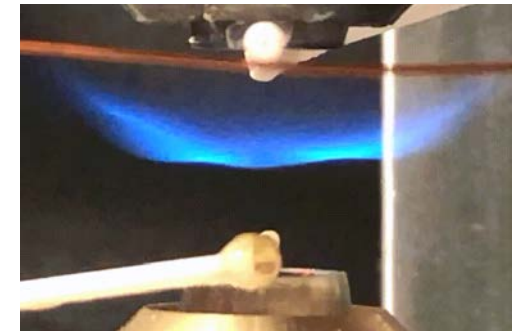
No discharge



n=0

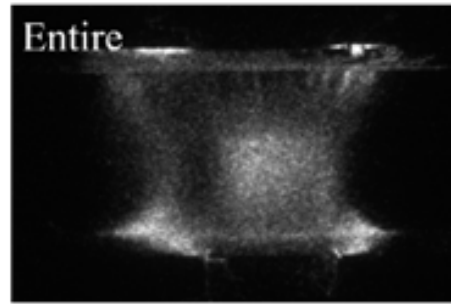
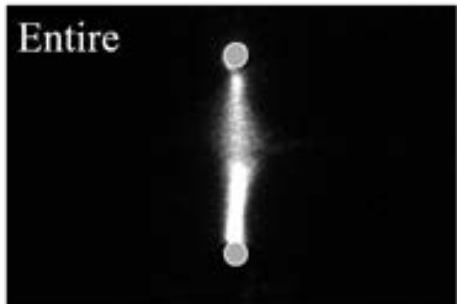
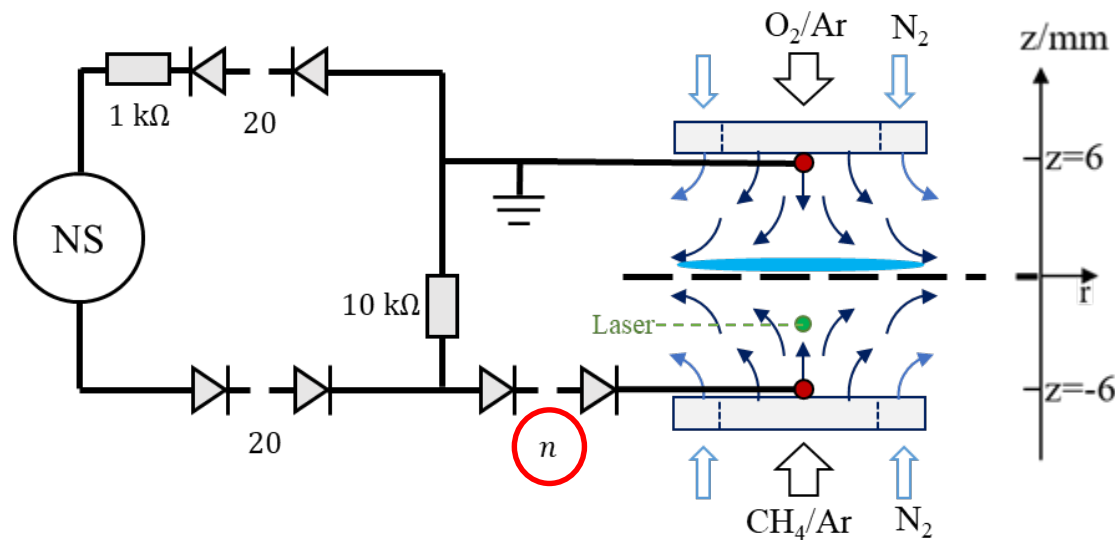


n=2

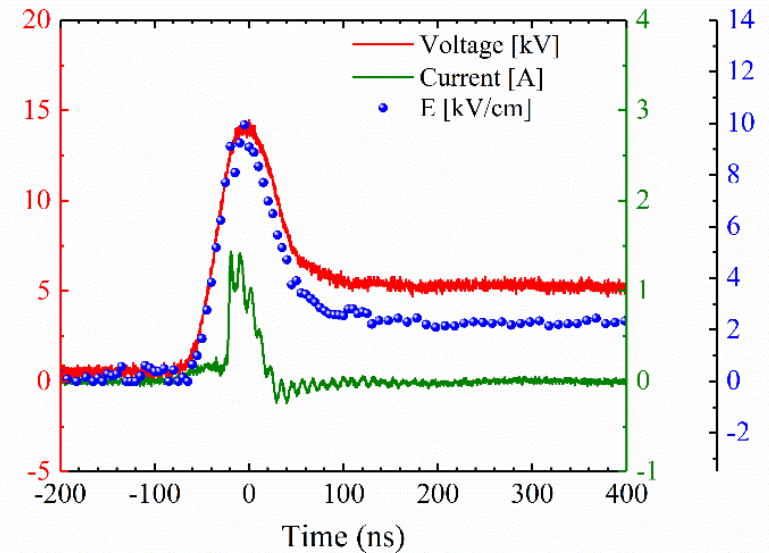


n=6

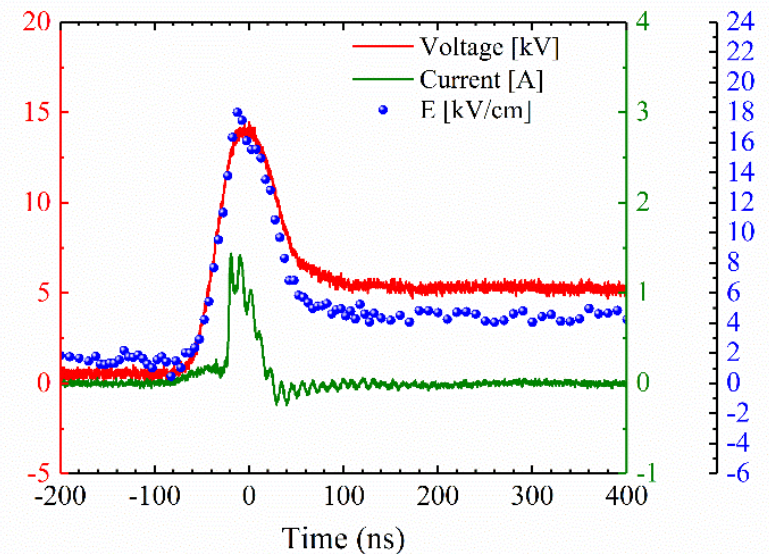
Ns Pulse Discharge Forces Flame Oscillations



n=4, z=1 mm



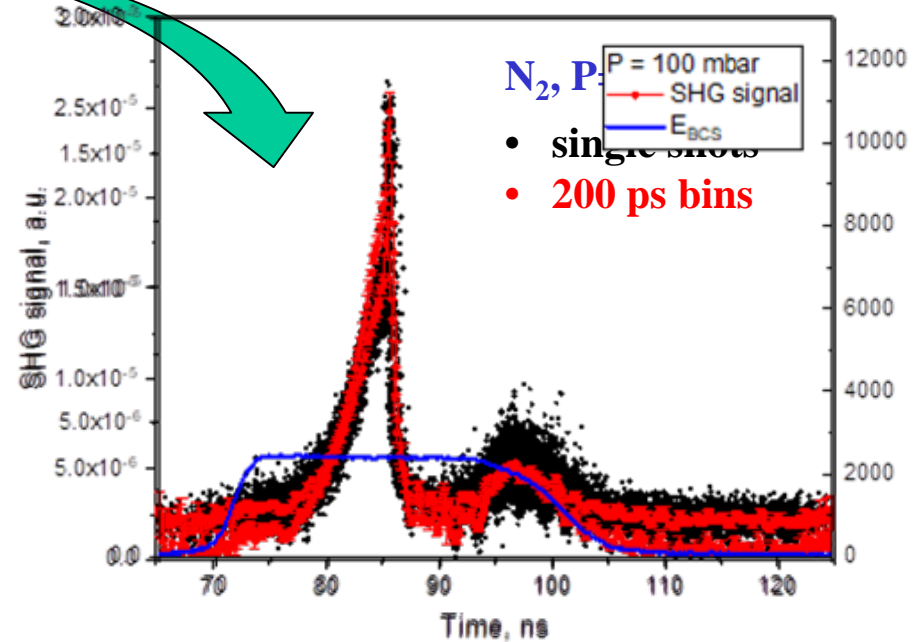
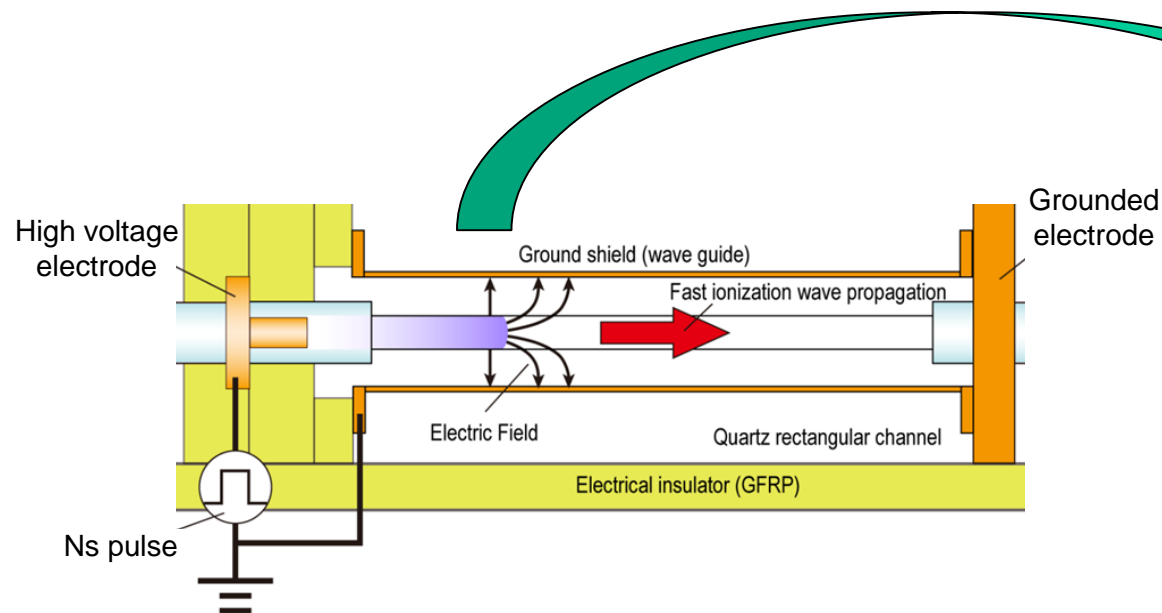
n=4, z=5 mm



- Ns pulse discharge generates ionization
- DC-like tail produces ion wind, moves the flame
- Potential for plasma “flameholder” development

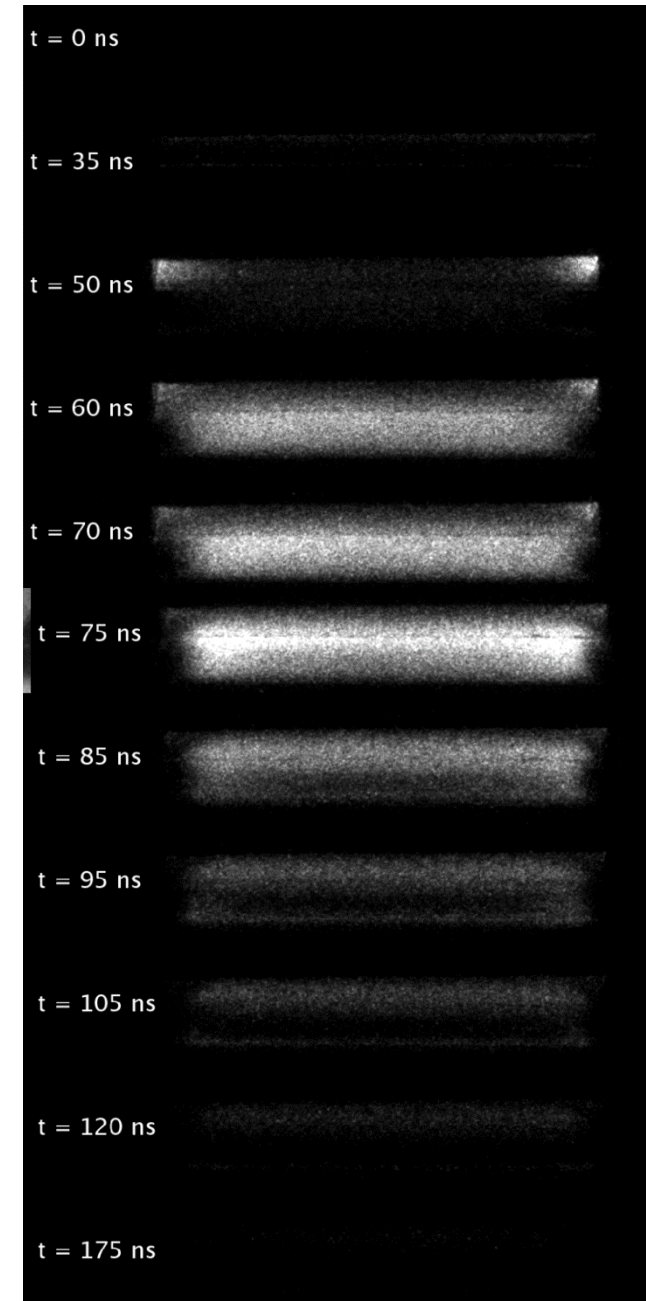
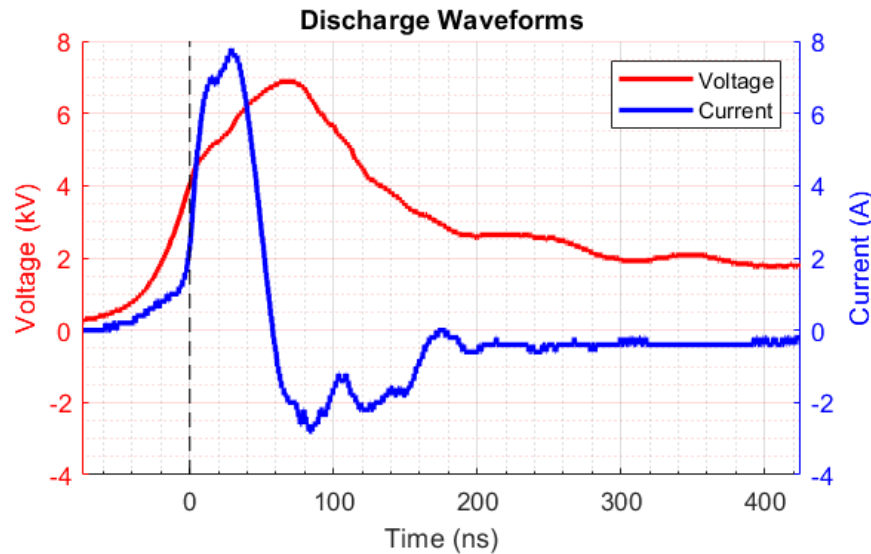
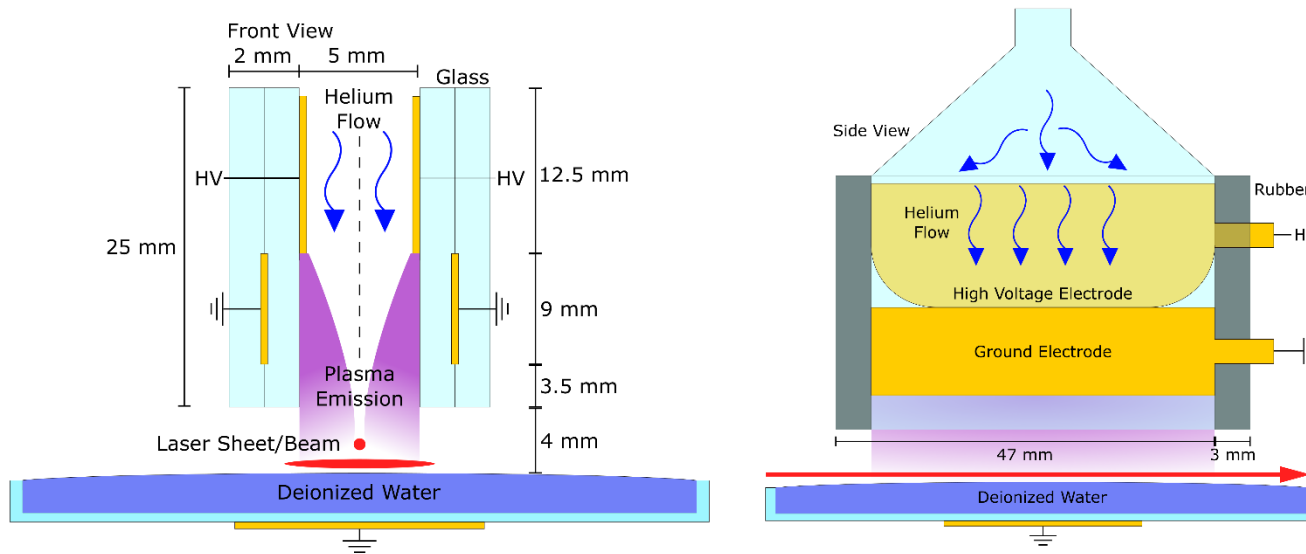
Ps E-FISH: Sub-ns Time Resolution

- Fast ionization waves (T.L. Chng, I. Orel, S. Starikovskaia, LPP): electric field profile across the wave front, time resolution 200 ps



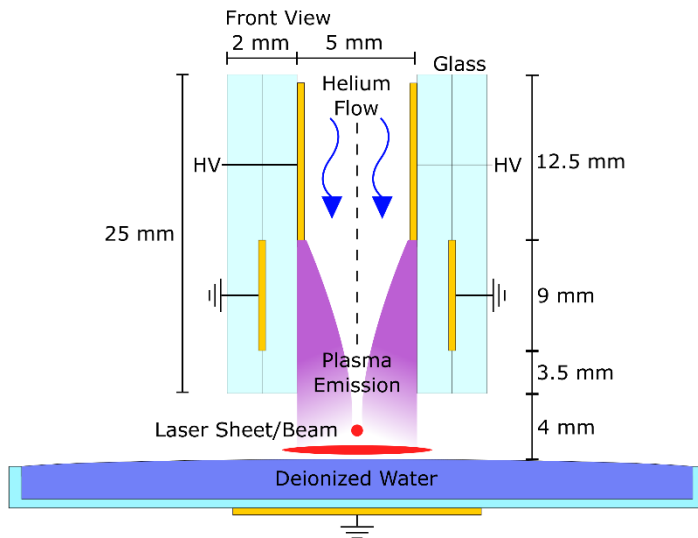
- Time resolution is limited only by laser pulse duration and scope resolution

2-D Atmospheric Pressure He Plasma Jet

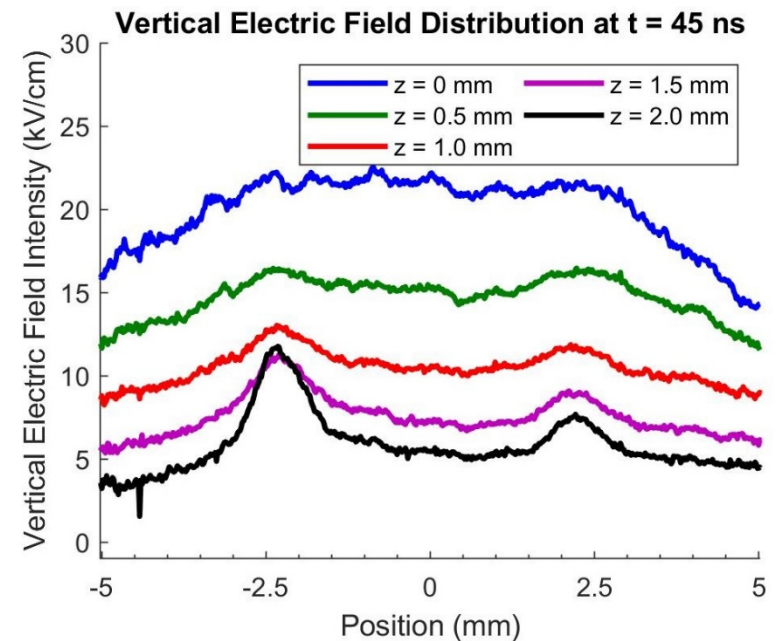
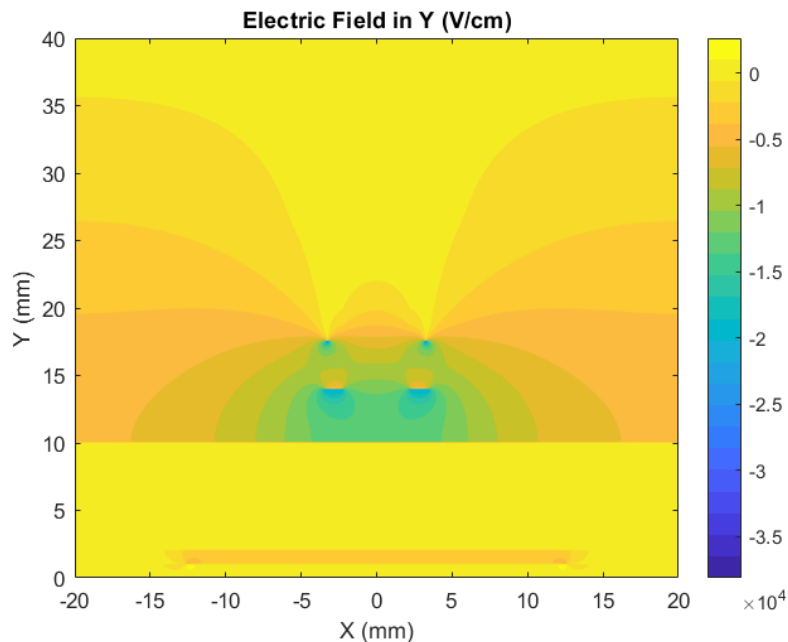
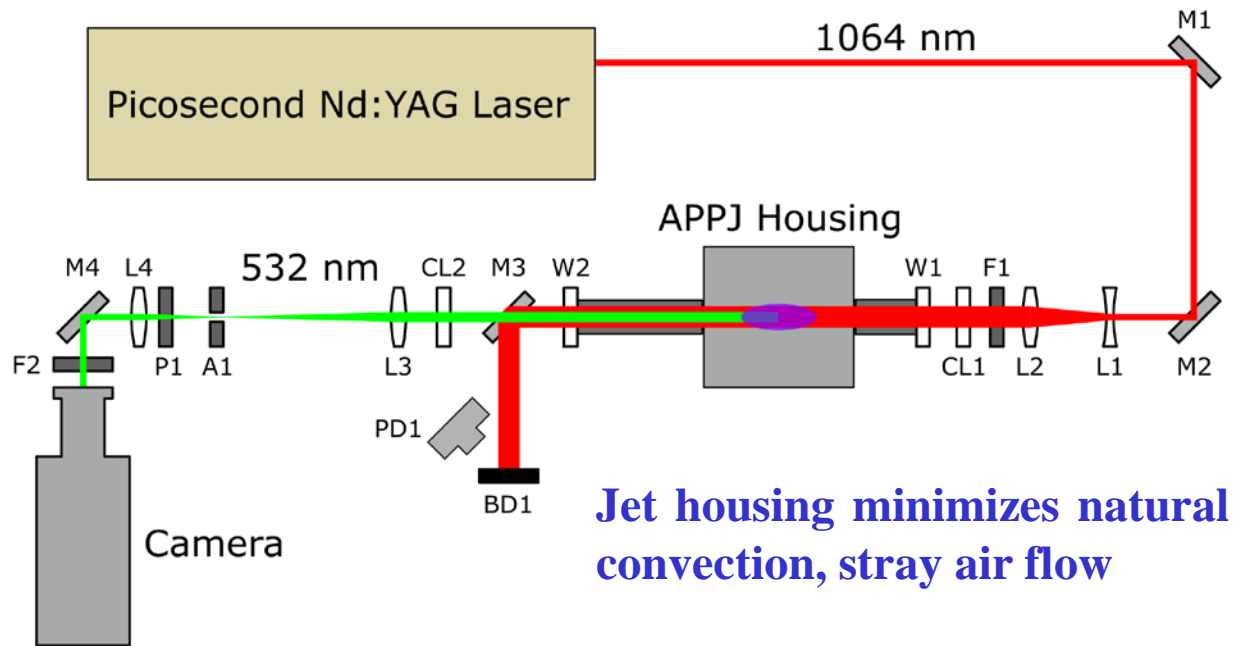


- He jet, discharge rep rate 10 Hz, negative polarity pulses

Results: 2-D Atmospheric Pressure He Plasma Jet

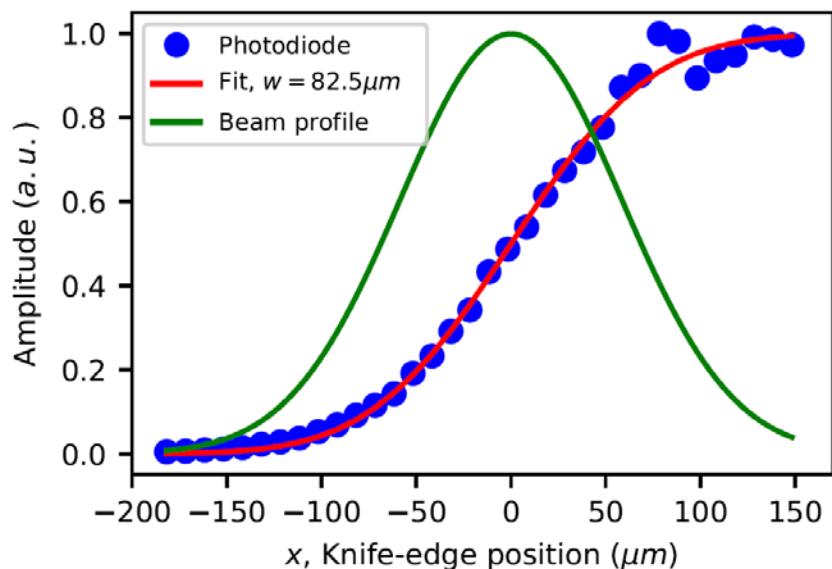
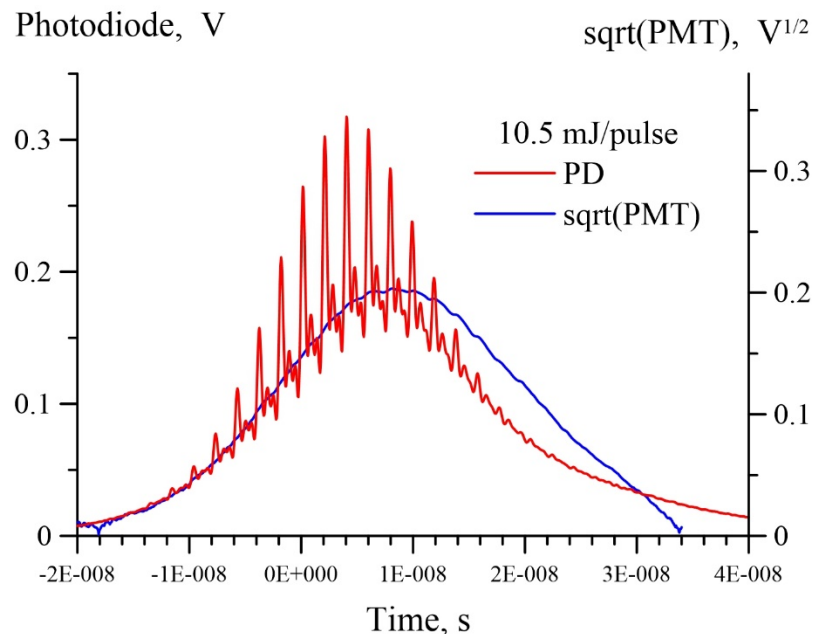
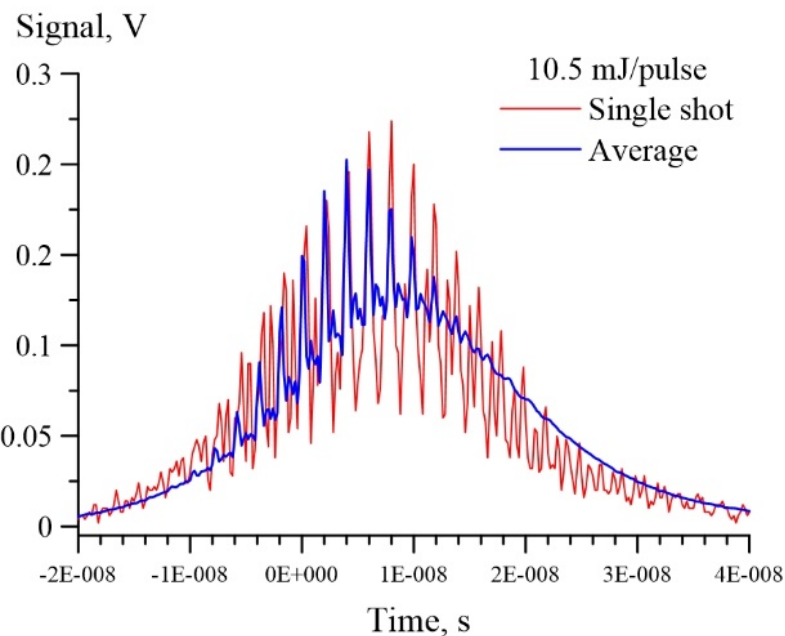


Calibration needs to be done in the same flow: cannot change geometry



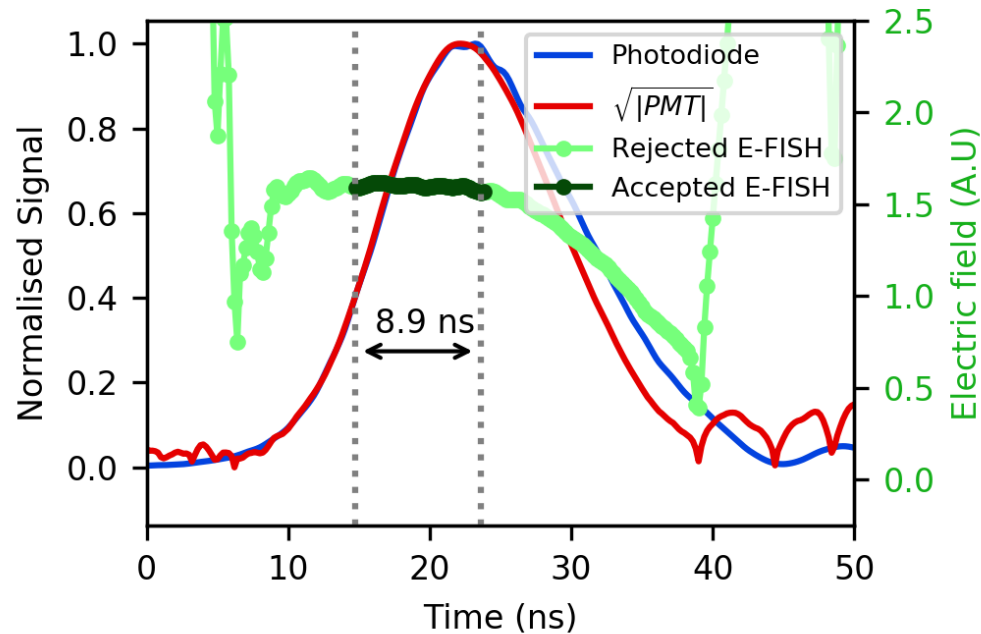
Ns EFISH: Is It Good for Anything?

T. Butterworth, T. Orriere, D. Pai, D. Lacoste, M.S. Cha @ KAUST

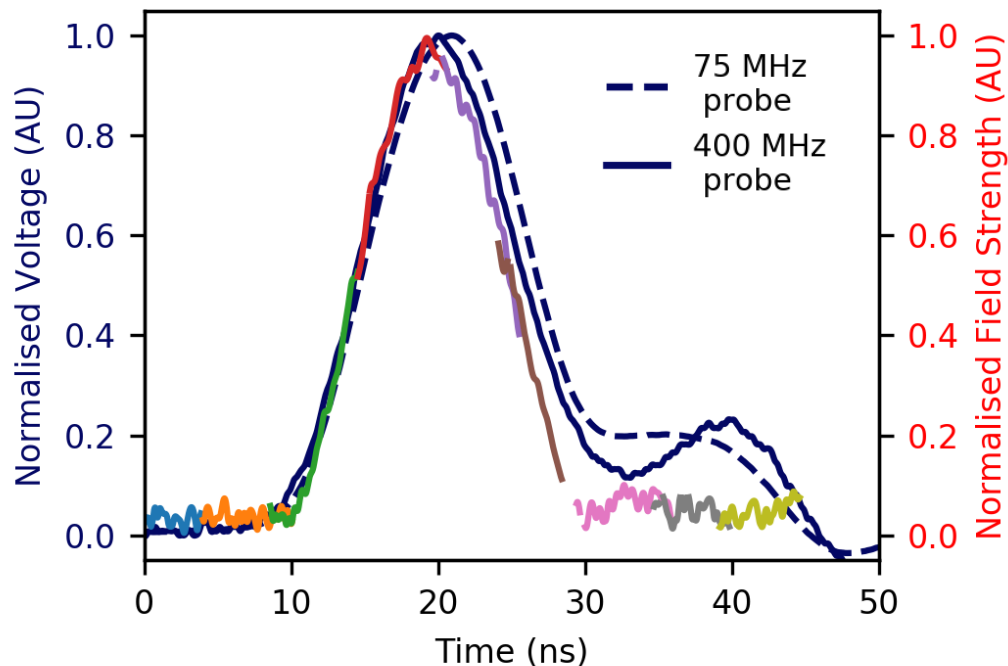


- “Bottom-shelf” Nd:YAG PIV laser, pulse FWHM 15-20 ns
- Pulse shape affected by mode beatings (≈ 0.5 ns, persistent at low pulse energies)
- Poor beam quality (FWHM 165 μm for $f=15$ cm lens, no breakdown even at 50 mJ/pulse)
- At constant field, $\text{sqrt}[\text{PMT}(t)] / \text{PD}(t) \neq \text{const}$
- There seems to be little hope. Give up?

Ns EFISH: Time-Accurate Laplacian Field

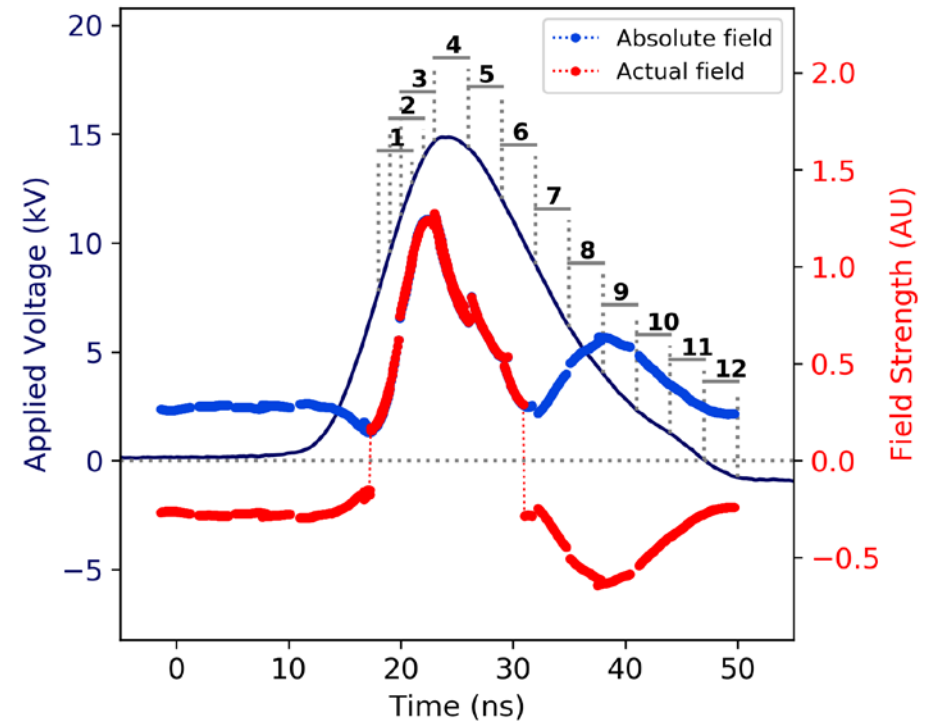
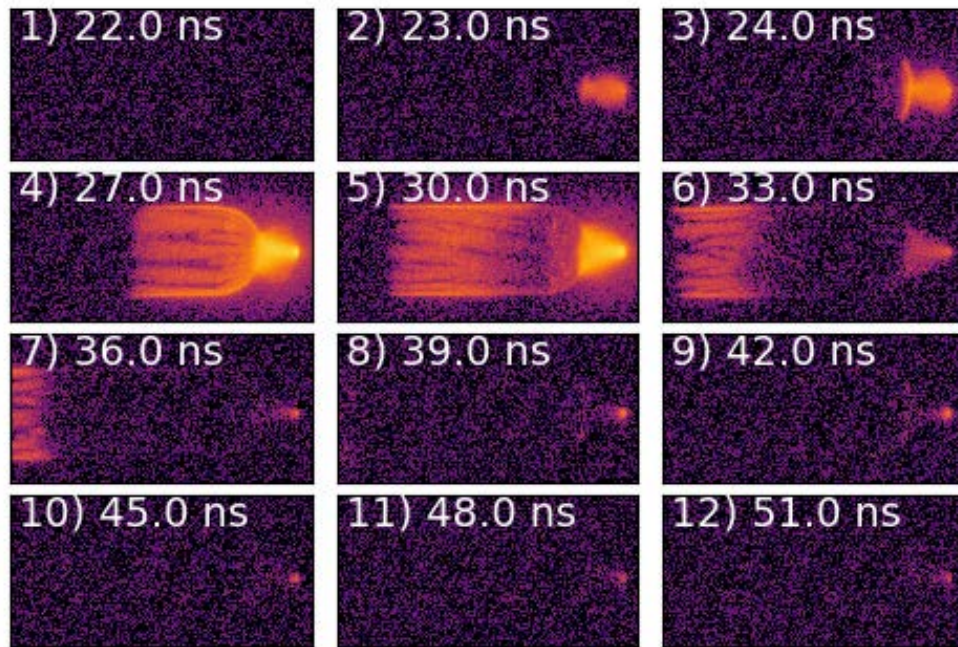


- Electric field constant in time
- Scope triggered on HV probe, laser pulse jitter “smears” the peaks
- Normalized $\sqrt{|PMT(t)|}$ and $PD(t)$ are on top of each other for half the pulse
- About 10 ns of accurate EFISH data



- Laplacian field pulse, FWHM 12 ns
- Air at 3 bar, to prevent corona formation near pin electrode
- $\sqrt{|PMT(t)|} / PD(t)$ data stitched together over multiple laser pulse delays 5 ns apart
- Very good agreement with electric field rise measured by high bandwidth probe

Ns EFISH: Data in Ns Pulse DBD Plasma



- Ambient air at 1 bar, pin-to-pin dielectric barrier discharge, electrode gap 2 mm
- Residual field due to charge accumulation on dielectric, from previous pulse
- Field reversal during both during voltage rise and fall
- Field is not zero during field reversal (averaged nearly uniformly over ≈ 1 cm)
- Absolute calibration from Laplacian field reversal timing (underway)

Summary

- Electric field in atmospheric pressure plasmas and flames measured by ps Electric Field Induced Second Harmonic (E-FISH)
- Ps E-FISH: simple, species-independent, 2 orders of magnitude more sensitive compared to CARS-like 4-wave mixing
- Sub-ns time resolution, measurements of individual electric field vector components
- Ns pulse plasma actuators: reduced electric field (E/N) controls rate of energy thermalization (temperature rise), effect on the flow via coherent structure formation
- Ns pulse discharge coupled with ms tail in counterflow flame: electric field forces strong flame oscillations, potential for plasma flameholding
- Atmospheric pressure helium plasma jet impinging on water: measurements of 1-D electric field distributions, insight into excited species and radicals generation
- Ns EFISH: electric field measurements on a time scale shorter than laser pulse

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- **Thomas Butterworth (KAUST), Thomas Orriere (KAUST), David Pai (U. Poitier), Deanna Lacoste (KAUST), Minsuk Cha (KAUST)**

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- **NSF “Fundamental Studies of Accelerated Low Temperature Combustion Kinetics by Nonequilibrium Plasmas”**
- **US DOE PSAAP-2 Center “Exascale Simulation of Plasma-Coupled Combustion”**