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

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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

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

(4) $N_2$ vibrational excitation:
Low reactivity, slow thermalization

Pulsed discharges (NS, AC DBD,): high $E/N$

(5,6) $N_2$, $O_2$ 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

\[ N_2 \text{ vibrational excitation: } N_2(v = 0) + e \rightarrow N_2(v > 0) + e \]

\[ N_2 \text{ electronic excitation: } N_2(X^1\Sigma) + e \rightarrow N_2(A^3\Sigma, \ldots) + e \]

\[ \text{O atom generation: } N_2(A^3\Sigma) + O_2 \rightarrow N_2(X^1\Sigma) + O + O \]

\[ \text{NO formation: } N_2(A^3\Sigma, \ldots) + O \rightarrow NO + N \]

Burnette et al, PSST, 2014
Shkurenkov et al, PSST, 2014
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 $u=96 \text{ m/s}$
Plasma / CFD Modeling Predictions
Baseline and forced flows, \( Re = 1.2 \cdot 10^6 \)

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, $E^{\text{ext}}$, 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 $2\omega$.

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

$I^{(2\omega)}$ – SH signal; \hspace{1em} $\chi^{(3)}$ – N – nonlinear susceptibility;
$L$ – interaction length (controlled by confocal parameter of the lens)

Phase matching parameter $\Delta k$ 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

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

- 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
• 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 😐

Simeni Simeni et al., Comb. Flame, 2018
More Interesting Case: Counterflow Flame

Counterflow, atmospheric pressure CH$_4$-O$_2$-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 generates ionization

DC-like tail produces ion wind, moves the flame

Potential for plasma “flameholder” development

Tang et al., Comb. Flame, 2019
Ps E-FISH: Sub-ns Time Resolution

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

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

Chng et al., PSST, 2019
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

Jet housing minimizes natural convection, stray air flow
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, \( \sqrt{\text{PMT}(t)} / \text{PD}(t) \neq \text{const} \)
- 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 $\sqrt{\text{PMT}(t)}$ 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
- $\sqrt{\text{PMT}(t)} / \text{PD}(t)$ 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
Acknowledgments

Students and Colleagues:

- Marien Simeni Simeni (OSU / U. Minnesota), Ben Goldberg (OSU / Princeton U.), Tang Yong (OSU / Tsinghua U.), Keegan Orr (OSU)
- Tat Loon Chng, Inna Orel, Svetlana Starikovskaia (Ecole Polytechnique, Paris)
- Thomas Butterworth (KAUST), Thomas Orriere (KAUST), David Pai (U. Poitier), Deanna Lacoste (KAUST), Minsuk Cha (KAUST)

Sponsors:

- US DOE Plasma Science Center “Predictive Control of Plasma Kinetics: Multi-Phase and Bounded Systems”
- NSF “Fundamental Studies of Accelerated Low Temperature Combustion Kinetics by Nonequilibrium Plasmas”
- US DOE PSAAP-2 Center “Exascale Simulation of Plasma-Coupled Combustion”