

Diagnostics capabilities at PCRF

Shurik Yatom and Arthur Dogariu



Overview I

1. Optical emission spectroscopy

Spectral imaging

Identification of excited species via broadband emission

High spectral and temporal emission analysis

Plasma characterization via line broadening /ratio

2. Laser induced fluorescence

What is LIF?

LIF measurements of atomic and molecular species

3. Laser Scattering : Raman\Thomson\Rayleigh Scattering

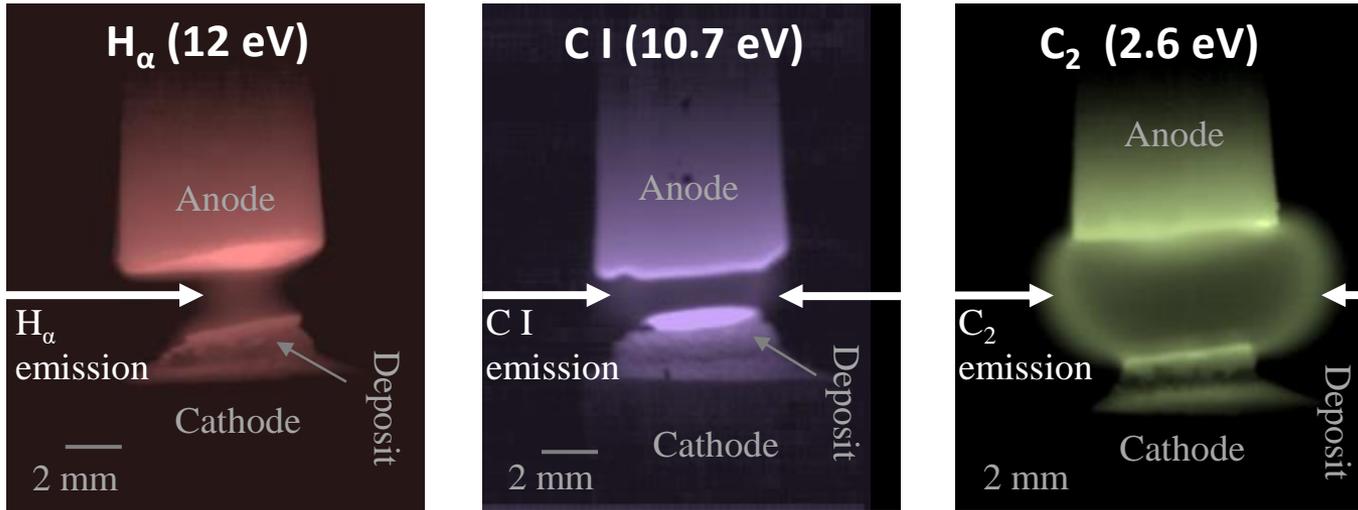
4. Laser-induced incandescence

5. Fourier Transform Absorption Spectroscopy (FTIR)

6. In development : CRDS

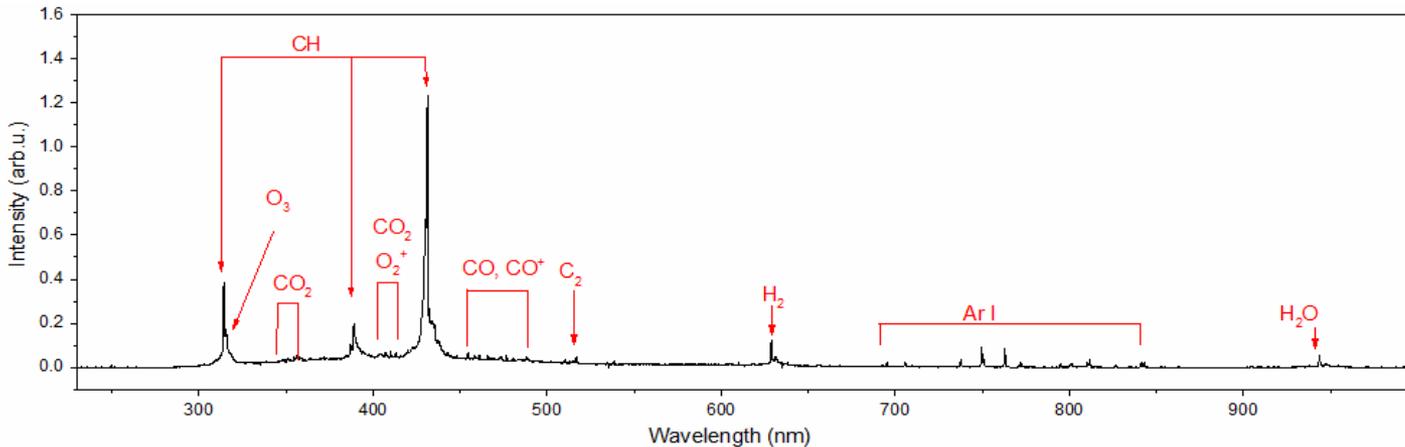
Optical emission spectroscopy

“Spectral” imaging of species in carbon arc*



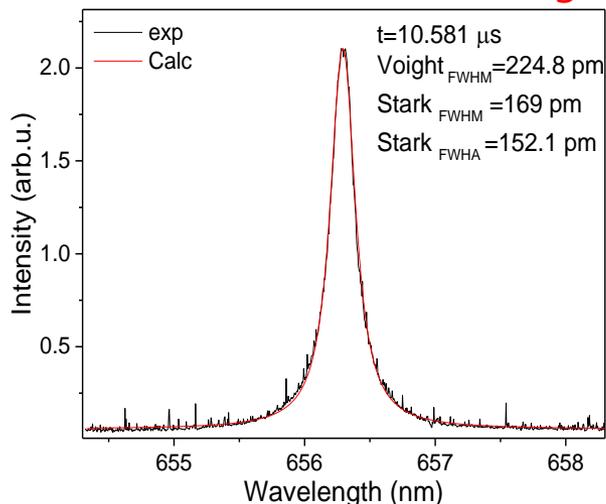
Identification of excited species via broadband emission
(plasma assisted gas conversion in DBD reactor)

Emission spectrum of plasma + Mn_2O_3 catalyst covered beads in $Ar+CH_4$ gas

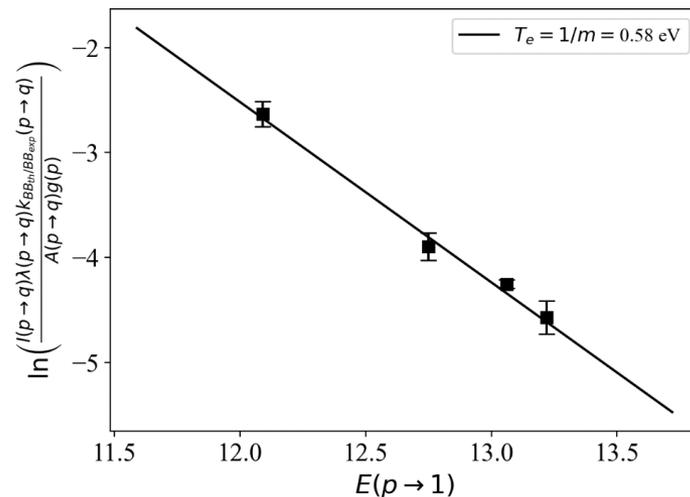


Plasma density and temperature via hydrogen Balmer series

H α profile and fit
Ne from Stark broadening

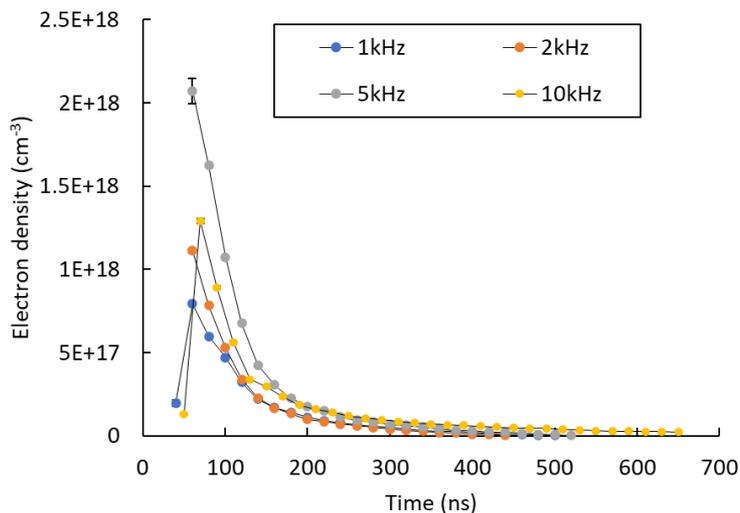


T_e from Boltzman plot of H-Balmer lines



Plasma density vs time in nanosecond discharge on thin liquid film PCRF project with Florida State University

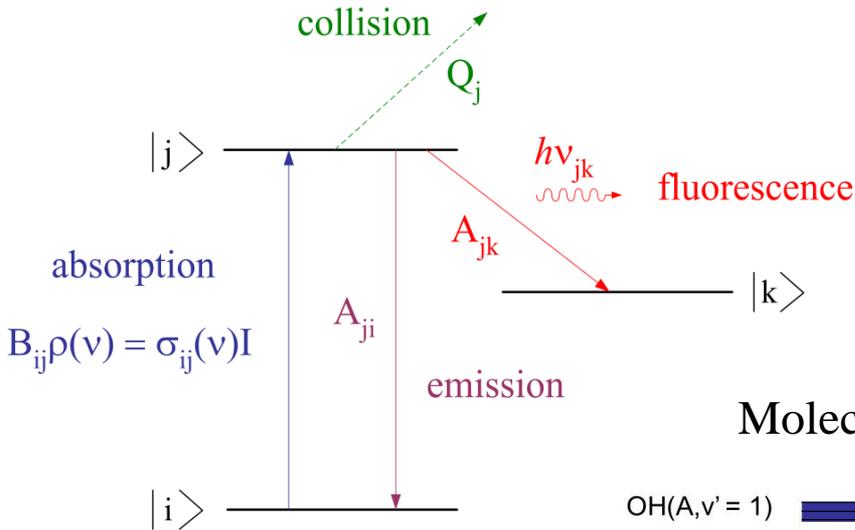
Role of Frequency (16kV, 40ns)



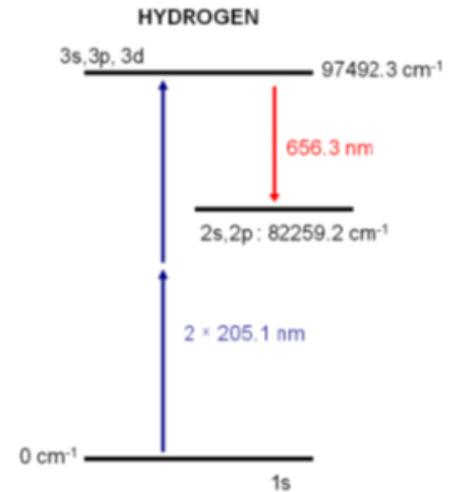
Laser-induced fluorescence (LIF)

Detection threshold: $n_e \geq 10^8 \text{ cm}^{-3}$
Spatial resolution $\sim 100 \mu\text{m}$
Temporal resolution $\sim 5\text{-}7 \text{ ns}$

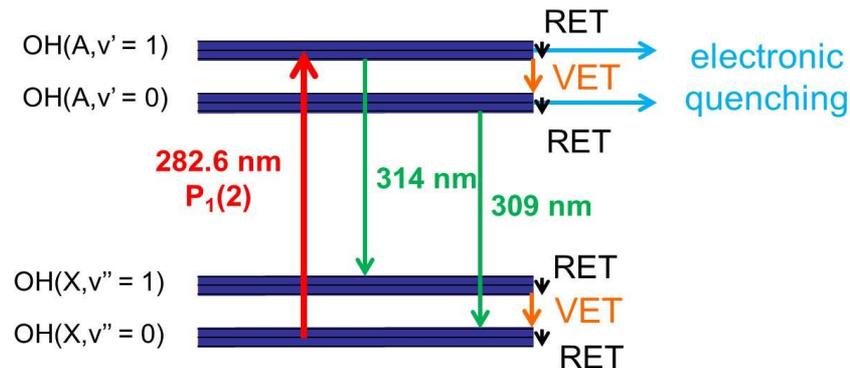
LIF scheme (atoms, ions)



TALIF= Two-photon LIF (light atoms)

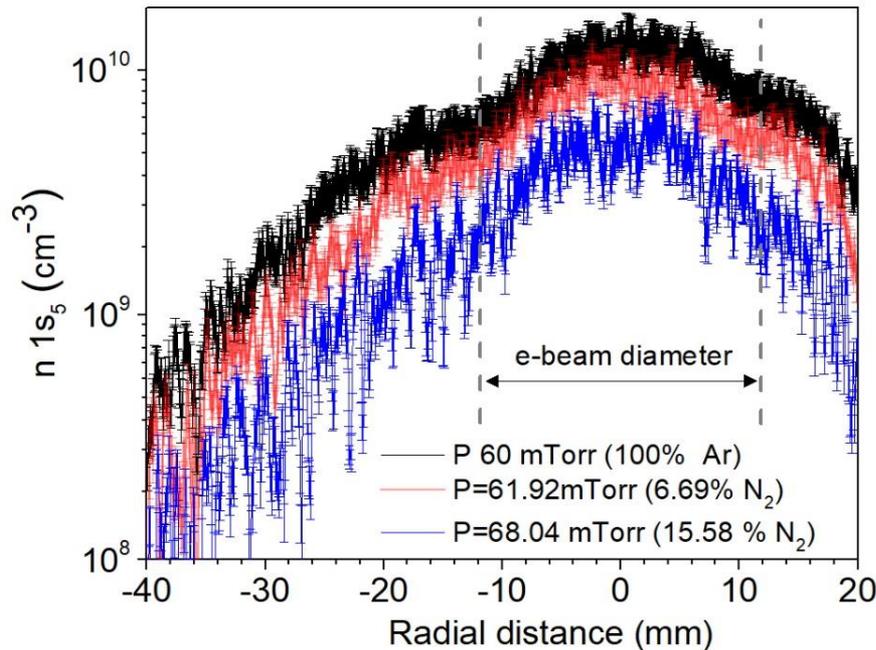
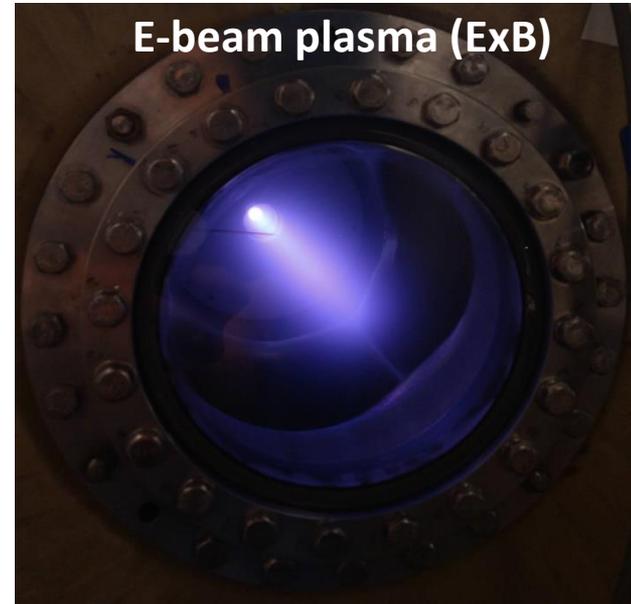
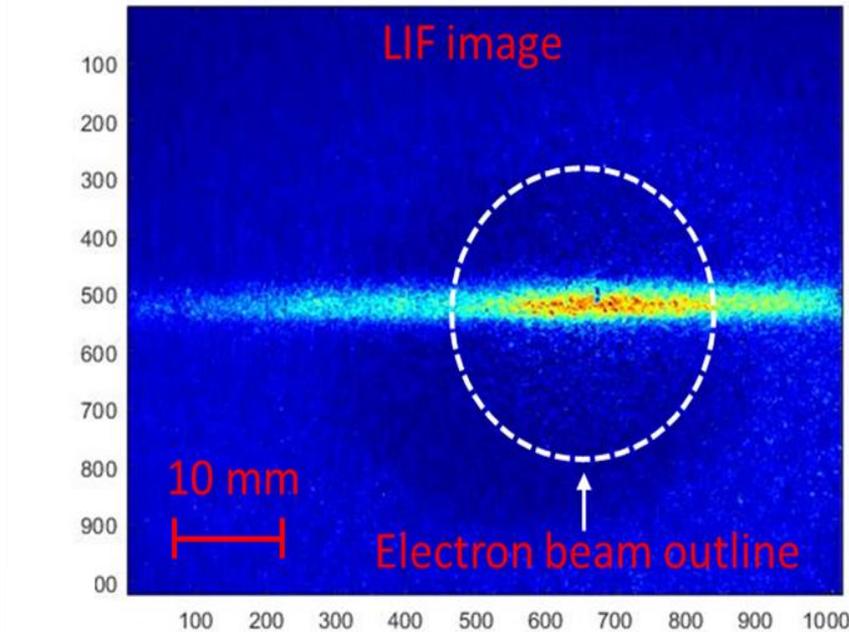


Molecules (LIF of OH)



rotational and vibrational energy transfer (RET, VET)
and electronic quenching

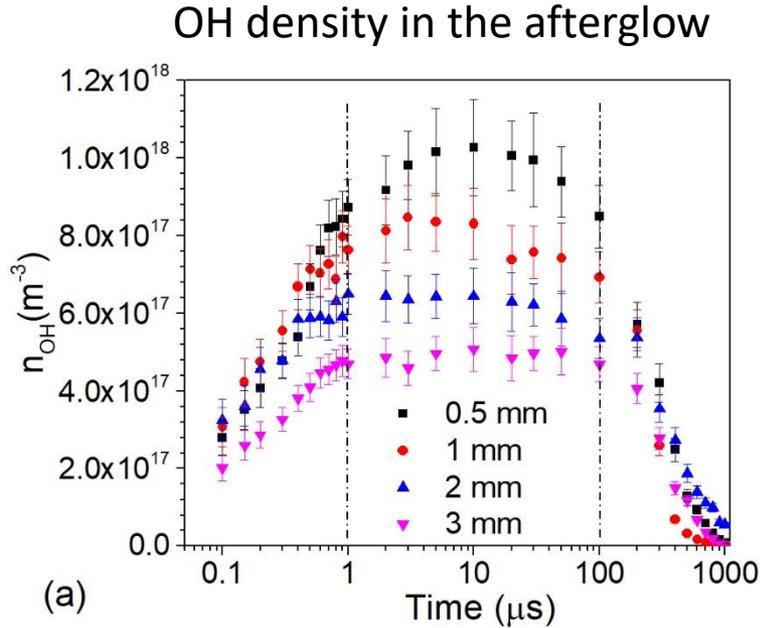
Ar 1s5 metastable species (PCRFB project with NRL)



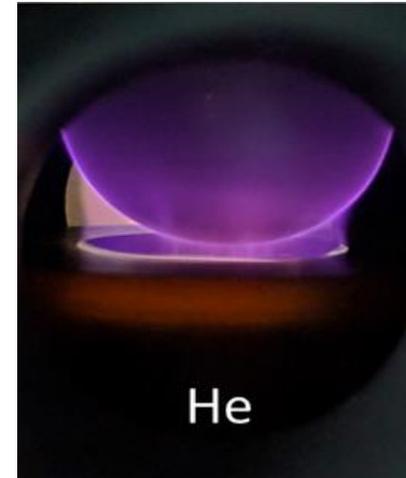
Spatio-temporal measurement of species (absolute density) (down to 10^8 cm^{-3})

LIF and PLIF on molecular species

OH production in ns DBD discharge (PCRF with Drexel U)

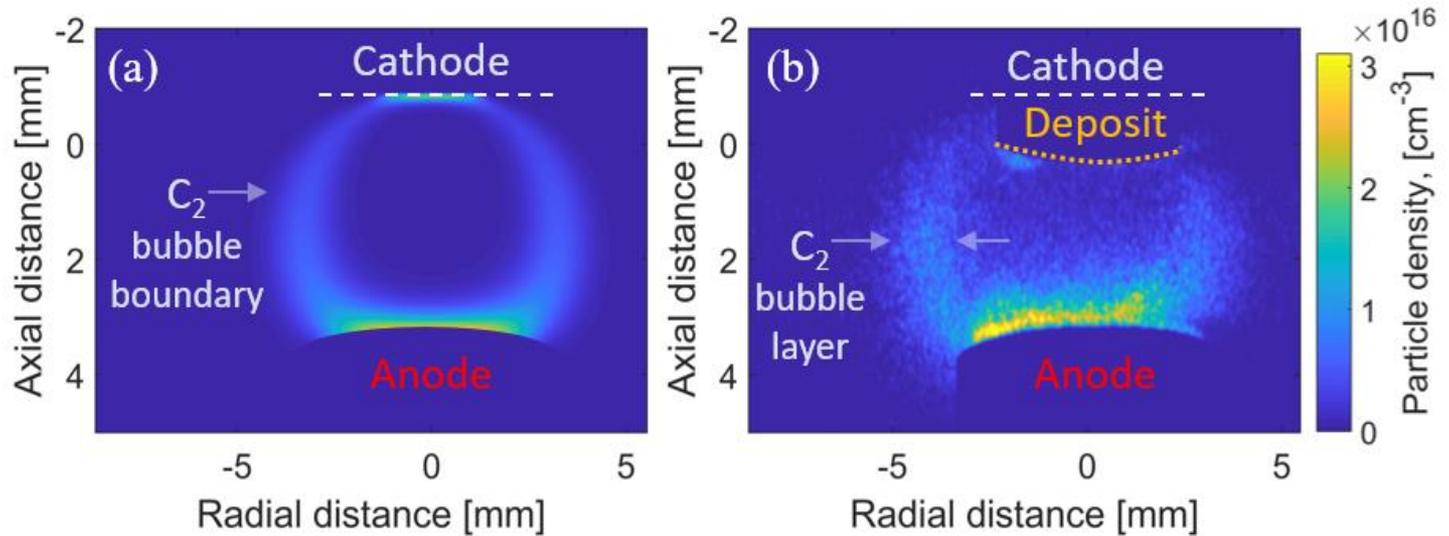


Floating DBD discharge in He



C₂ distribution in carbon arc : Planar LIF (PLIF) – quantitative imaging of species

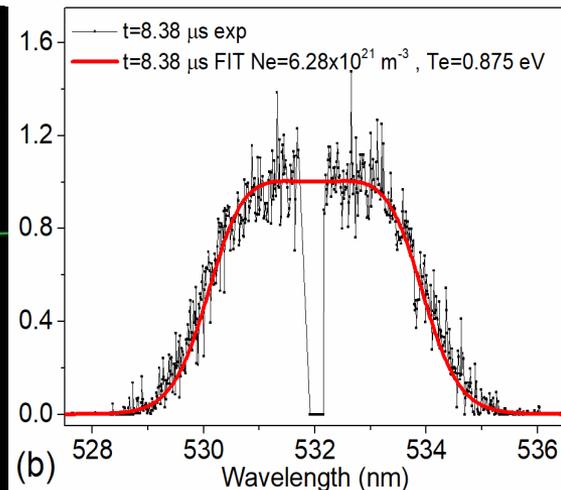
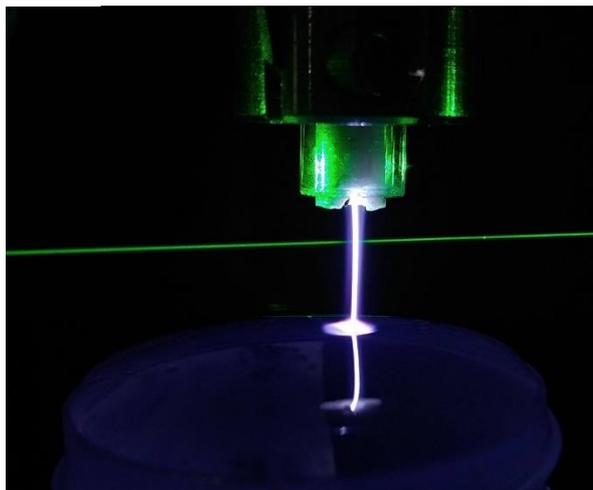
CFD (left) vs
Experiment
(Right)



Laser Scattering – Thomson, Raman and Rayleigh (PCRF project with Washington University at St. Louis)

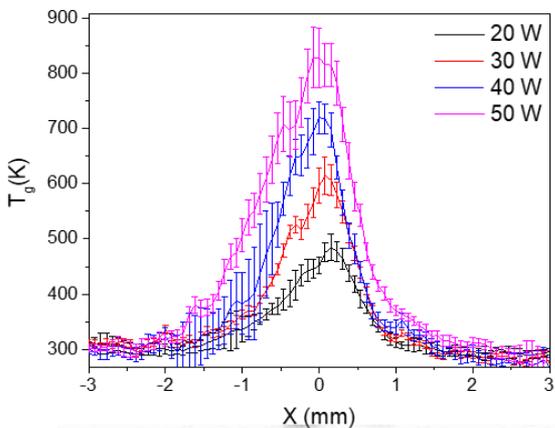
Detection threshold: $n_e \geq 10^9 \text{ cm}^{-3}$
Spatial resolution $\sim 200 \mu\text{m}$
Temporal resolution $\sim 20 \text{ ns}$

Thomson on RF-jet interacting with water

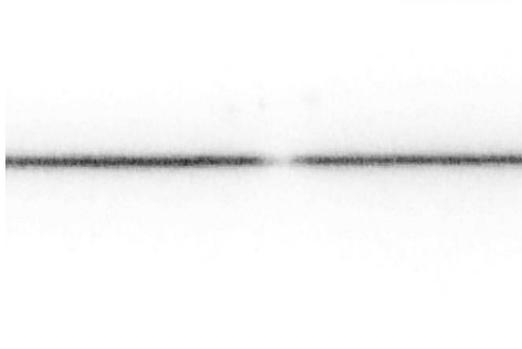


Rayleigh scattering

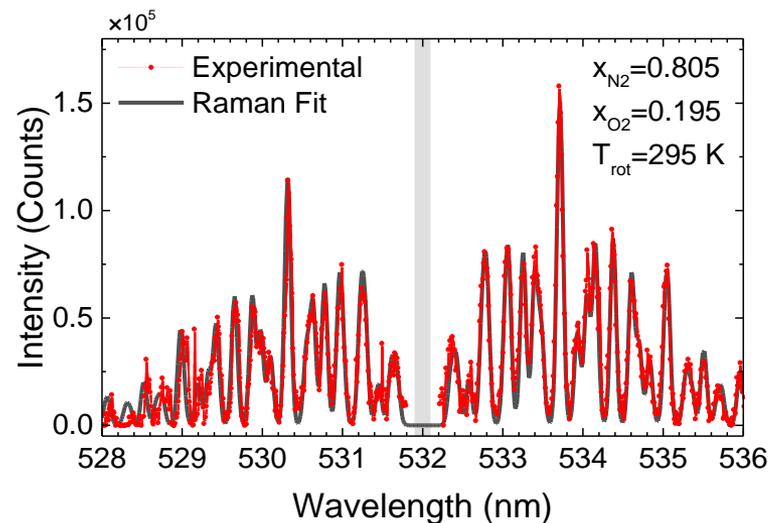
1D T_g profile



(b) Plasma on



Raman scattering



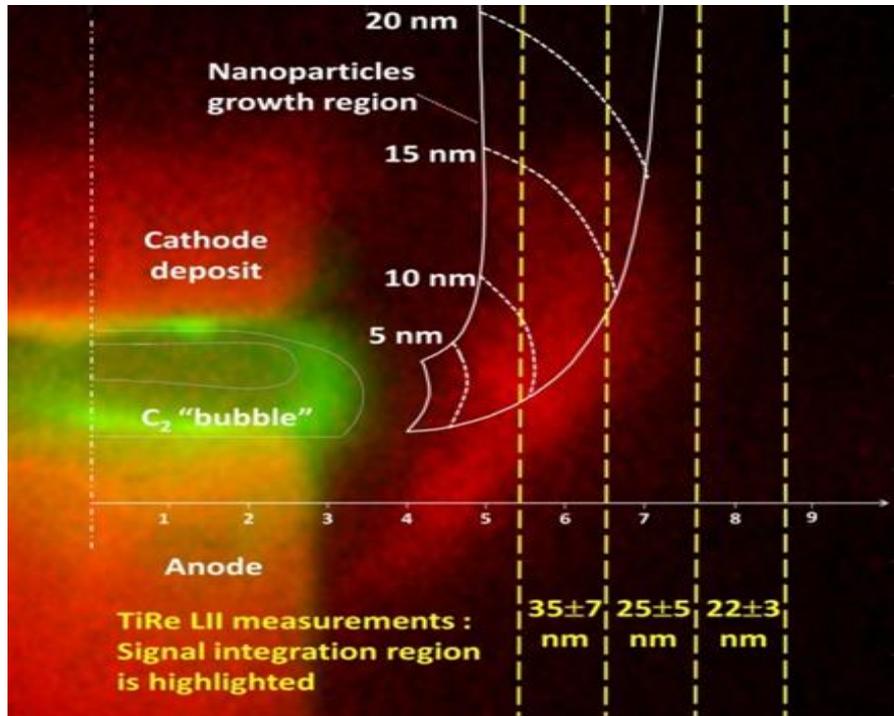
Laser Induced Incandescence (LII)

In-situ nanoparticle detection and characterization

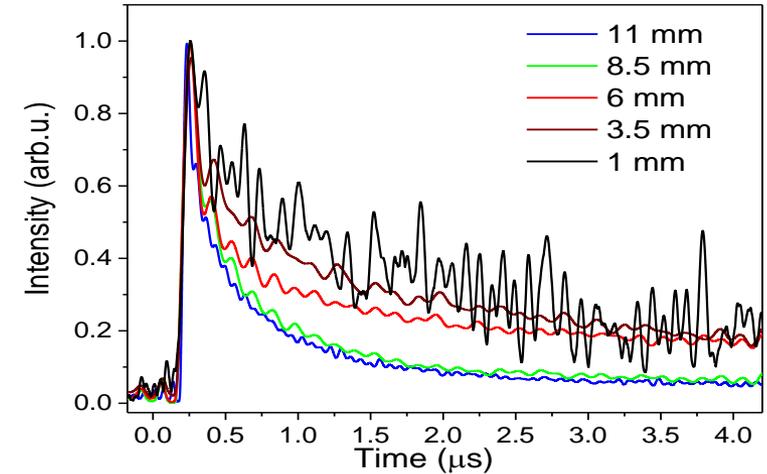
Particle size and density for volumetric particles ($D_p=10\text{ nm} - 5\text{ }\mu\text{m}$)

Spatial resolution 0.5-1 mm, Detection threshold: 10^8 cm^{-3}

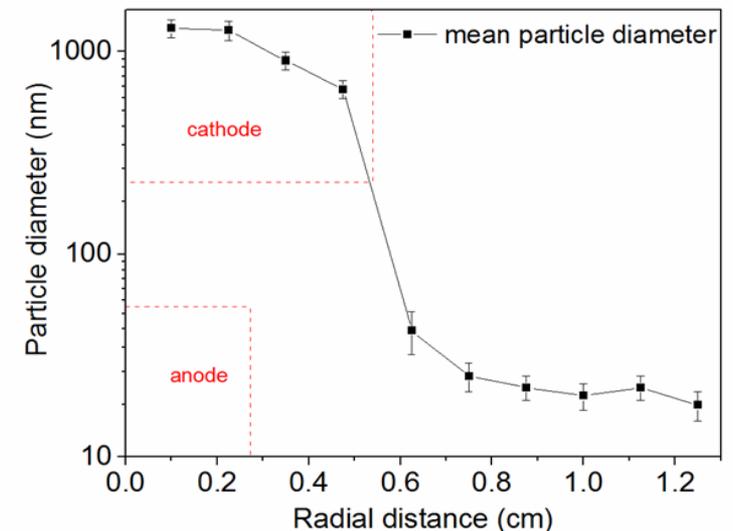
Planar LII imaging of nanoparticles/nanotubes



TiRe LII for different "r"

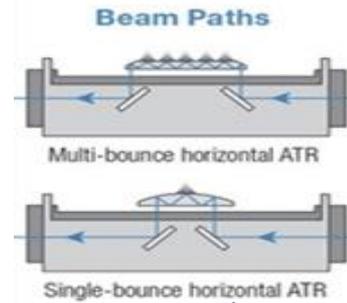
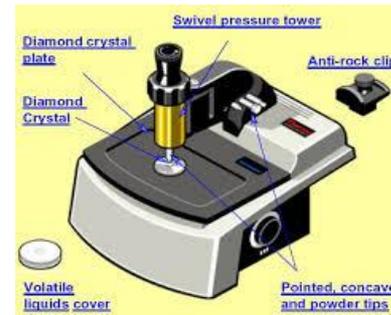


Mean particles diameter VS radial distance



FTIR : Fourier Transform Absorption Spectroscopy

- **JASCO FTIR-AS:** 7800 cm^{-1} - 400 cm^{-1} , scan time < 1 sec, max resolution 0.5 cm^{-1} , MCT: LN₂ cooled detector, low noise, high sensitivity - to 50 ppm with a 10 cm path length



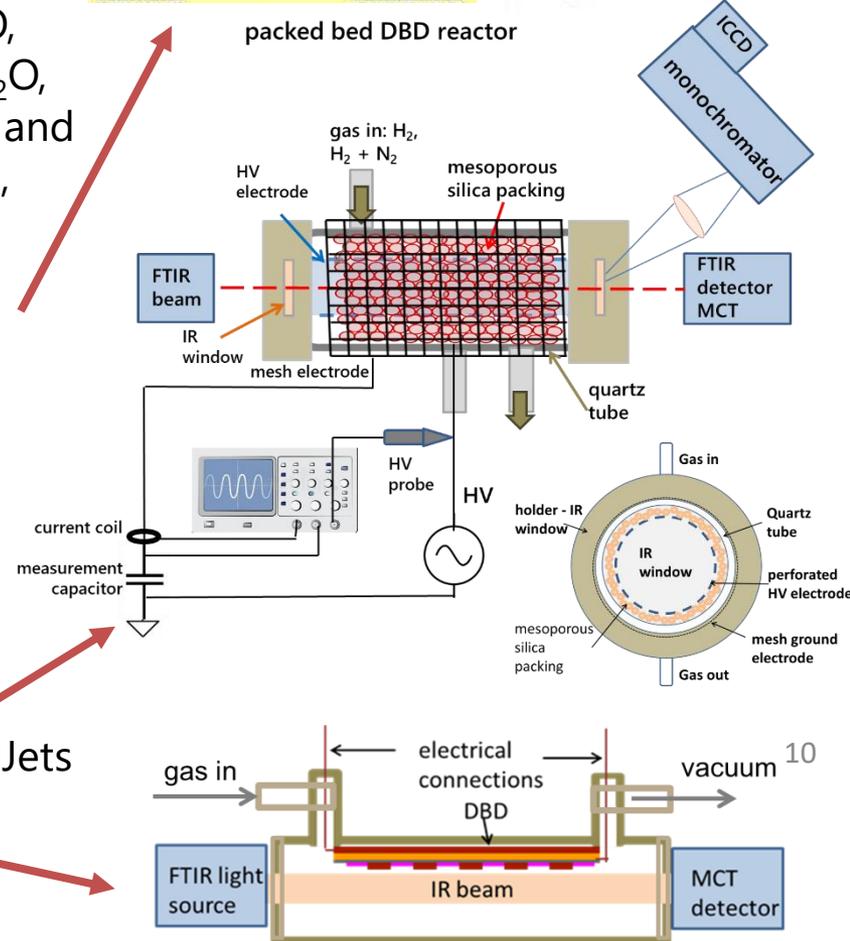
- Gas phase diagnostics: absolute concentrations for molecular species with active dipole modes: CO, CO₂, H₂O, CH₄, other hydrocarbons, NO, NO₂, N₂O, NH₃, O₃, OH, semiconductor NF₃, SiF₄, CF₄, CO₂, and many chloro/fluorocarbons, alcohols, aldehydes, and aromatic compounds

- **Attenuated Total Reflectance (ATR):** for long-life surface modification - plasma/surface interaction

- Range 10,000 - 300 cm^{-1}
- Samples as small as 50 - 100 μm

- **Additional Capabilities: In situ and in operando:**

- Packed bed or other **type reactor with center optical path**
- **Gas cell for in-situ testing** of surface DBD and Jets



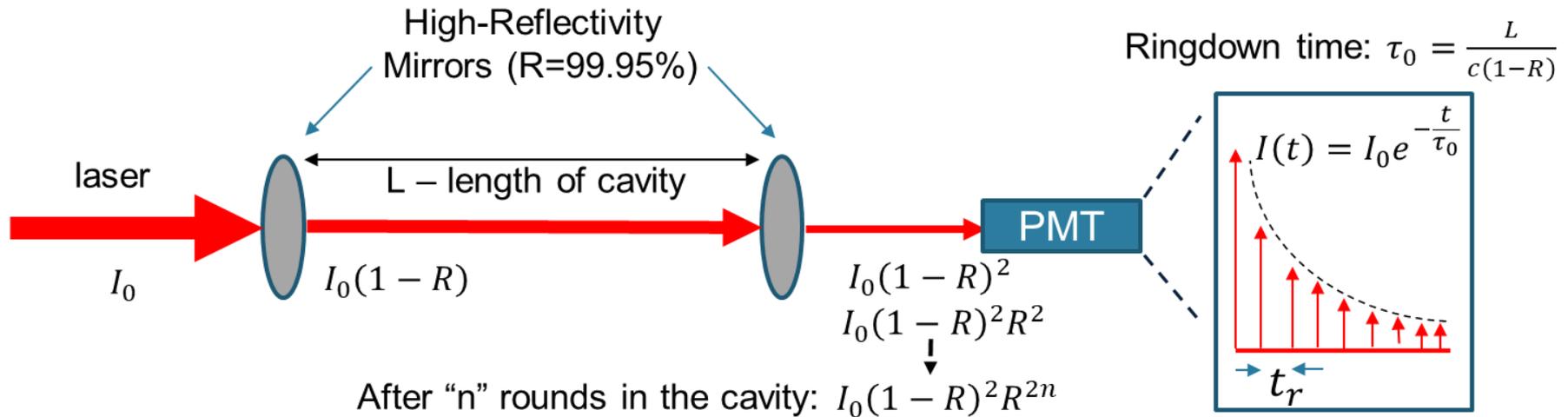
New diagnostics in development

Cavity ring-down spectroscopy (CRDS)

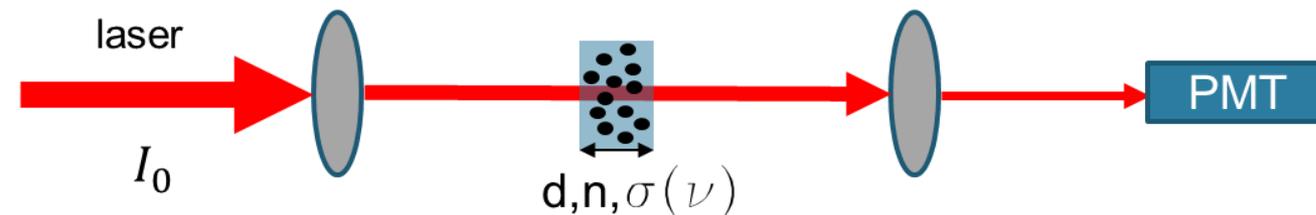
Absolute density measurement for atomic and molecular species

Doesn't depend on measurement of fluorescence decay time – applicable in very collisional environments

Doesn't require LIF transition scheme – applicable for species without known LIF scheme (CH₃)



If cavity has an absorber:



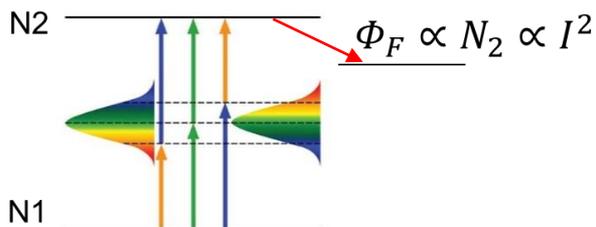
When absorber is present with absorbance $A = d \cdot n \cdot \sigma(\nu) = \frac{L}{c} \left(\frac{1}{\tau_{eff}} - \frac{1}{\tau_0} \right)$

Overview II

- 1. Two-Photon Laser Induced Fluorescence (TALIF)**
 - Density measurements and mapping of atomic species
 - High spatial and temporal resolution
 - High sensitivity achieved with low fluence
- 2. Hybrid fs/ps Coherent Anti-Stokes Raman Scattering (CARS)**
 - Single shot vibrational/rotational spectroscopy at 1kHz
 - Non-equilibrium temperature measurements
 - Species measurements – trace capability
- 3. Electric Field Induced Second Harmonic (E-FISH)**
 - Non-intrusive E-field measurements and mapping
 - High spatial and temporal resolution
- 4. Femtosecond Laser Electronic Excitation Tagging (FLEET)**
 - Non-intrusive velocimetry – flow mapping
- 5. Radar Resonantly Enhanced Multi-Photon Ionization (Radar-REMPI)**
 - Electron density with sub-ns resolution

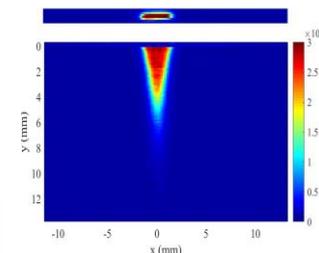
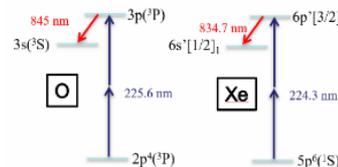
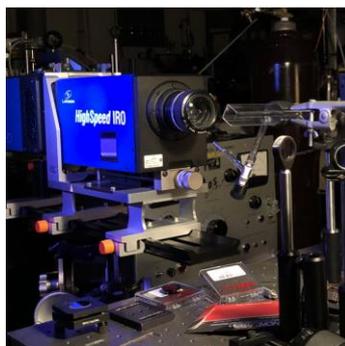
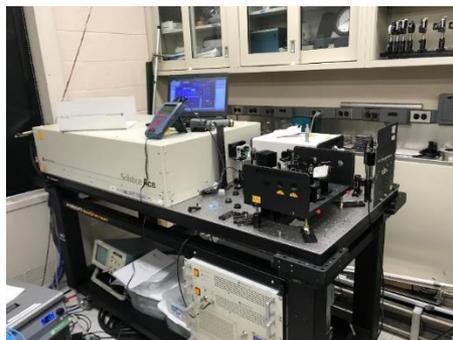
FS-TALIF – Two-photon Absorption Laser Induced Fluorescence

Femtosecond Two-Photon Absorption Laser Induced Fluorescence (FS-TALIF)



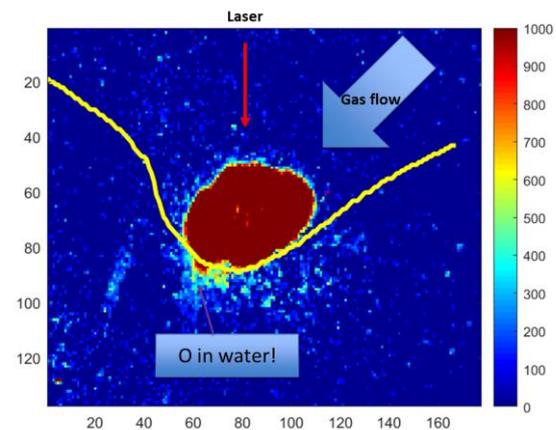
Advantages of ultrashort pulse:

- Easy to achieve two-photon transitions
- Reduced energy/pulse
- No collisions during excitation
- Easy resonance tunability
- No Doppler effects



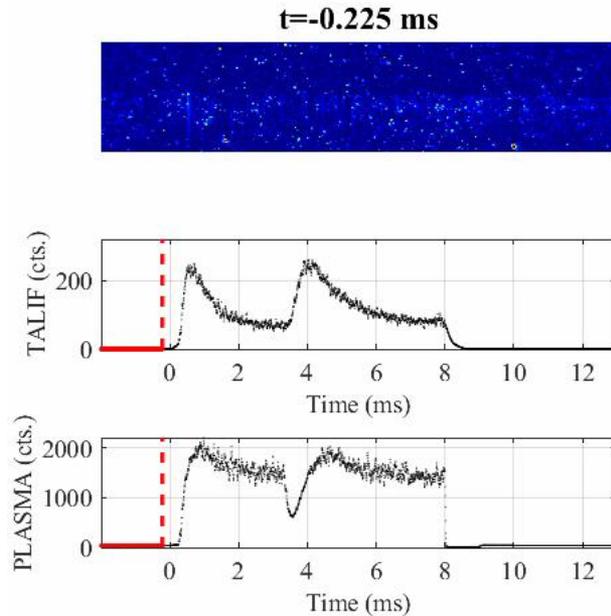
O-TALIF atomic oxygen generated by the COST plasma jet measured in air and in liquid

Imaging solvated plasma generated O atoms in water



with NCSU (PI: Katharina Stapelmann)

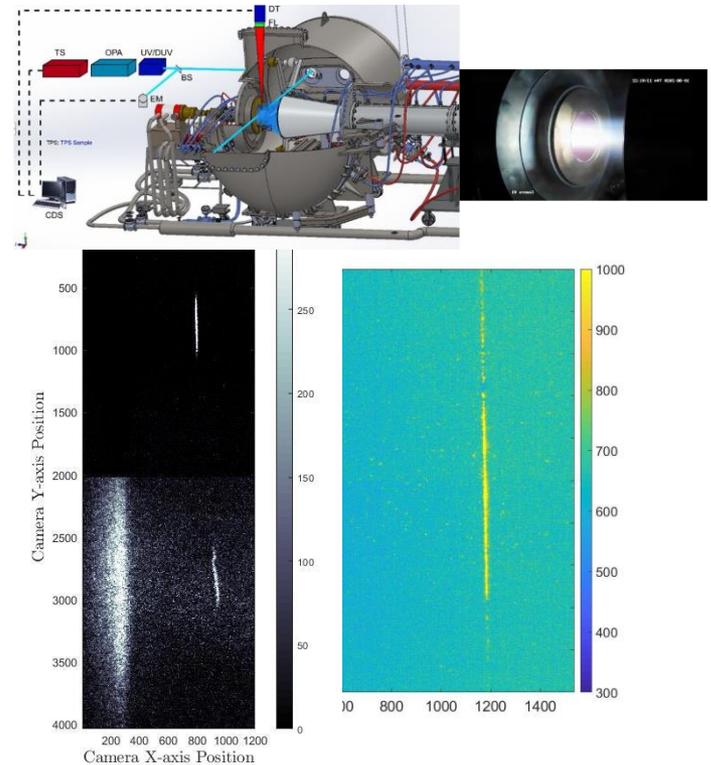
Femtosecond Two-Photon Absorption Laser Induced Fluorescence (Fs-TALIF)



- FRC (Field Reversal Configuration) RF heated magnetized cylindrical plasma mirror device
- Non-invasive measurements of neutral H concentration, dynamics of production and depletion under steady state and pulsed RF plasma
- Densities $\sim 10^{10} \text{cm}^{-3}$ measured using fs-TALIF

A. Dogariu, S.A. Cohen, P. Jandovitz, S. Vinoth, E.S. Evans, and C.P.S. Swanson *Rev. Sci. Instrum.*, in print, (2022).

with PPPL (PI: Sam Cohen)



FLEET velocimetry (left) and O-TALIF imaging (right) during MACH 6 hypersonic arc jet plasma flow.

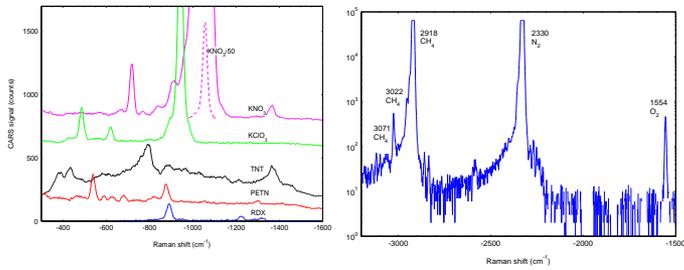
V. Gopal, D. Palmquist, L. Maddalena, L. E. Dogariu, and A. Dogariu, *Exp. Fluids* 62(10), 212 (2021).

with UT Arlington (PI: Luca Maddalena)

Fs/ps Hybrid CARS

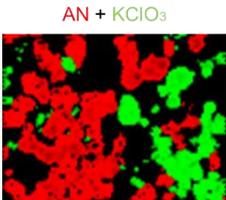
Coherent Anti-Stokes Raman Scattering (CARS) spectroscopy:
 Ultrafast diagnostic for gas/liquid/solid - characterization of atmospheric pressure plasmas and interfacial plasmas (plasma-liquids, plasma-solid state etc.)

- Gas density and temperature
- Solid/liquid molecular composition
- Surface changes under plasma interaction

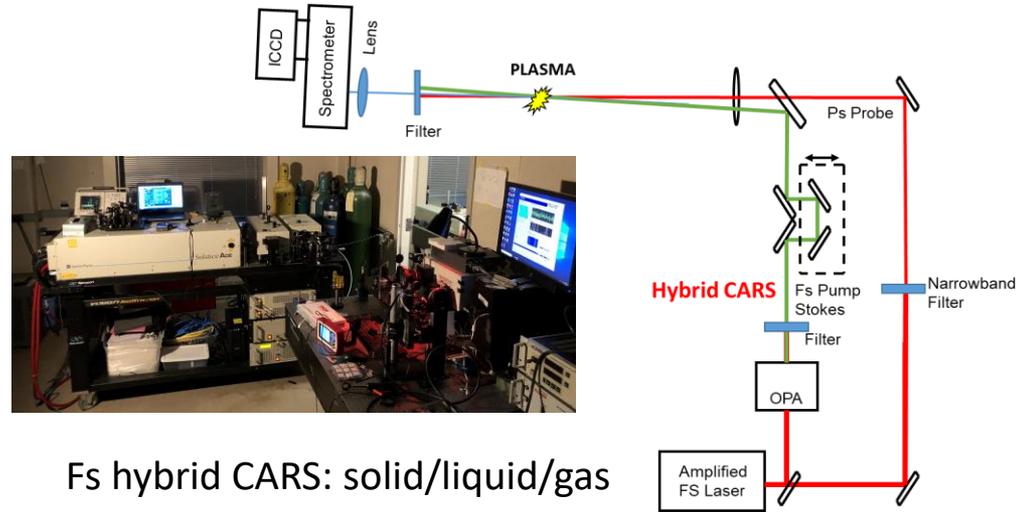


Backwards scattered CARS from solid/liquid surfaces

Forward hybrid CARS for gases



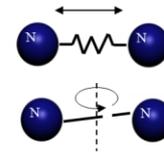
Species identification vibrational CARS



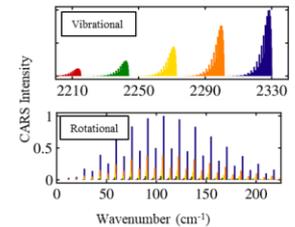
Fs hybrid CARS: solid/liquid/gas

CARS thermometry

$$N(v, J) \propto \exp\left(-\frac{E_{vib}(v)}{kT_{vib}}\right) \exp\left(-\frac{E_{rot}(J)}{kT_{rot}}\right)$$



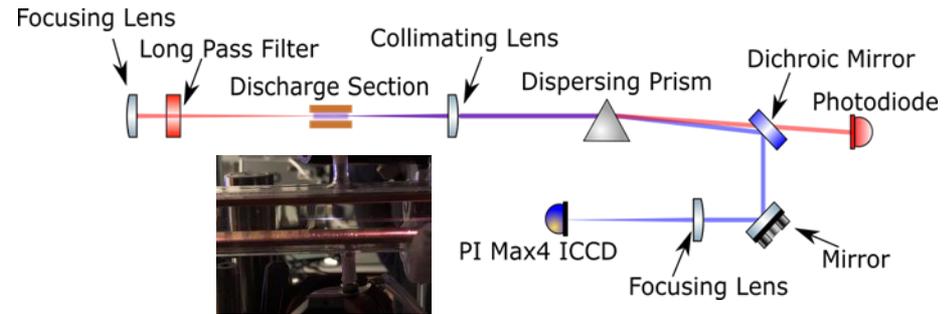
- Combustion
- Air vehicle propulsion systems
- Gas dynamics
- Plasma



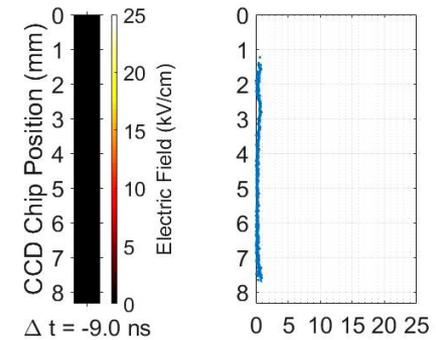
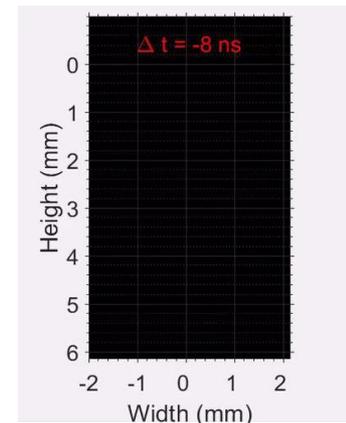
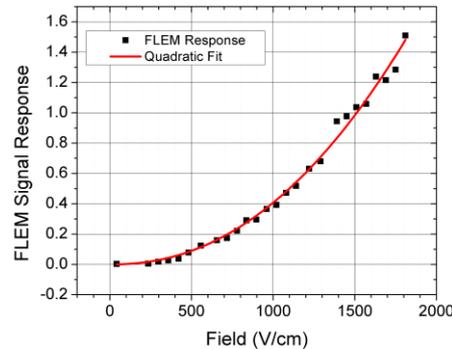
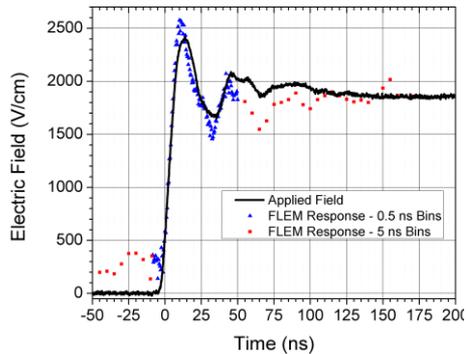
Electric-Field Induced Second Harmonic generation (E-FISH)

Fs laser based diagnostic for measuring E-field with high spatial and temporal resolution

1. Quadratic Dependence on E-Field
2. Time Resolution – femtosecond
3. Spatial Resolution – sub-mm
4. Species Independence – non-resonant, any gas/plasma
5. Field Vector Sensitivity – E field



E-FISH in an Atmospheric Pressure Plasma Jet (APPJ)

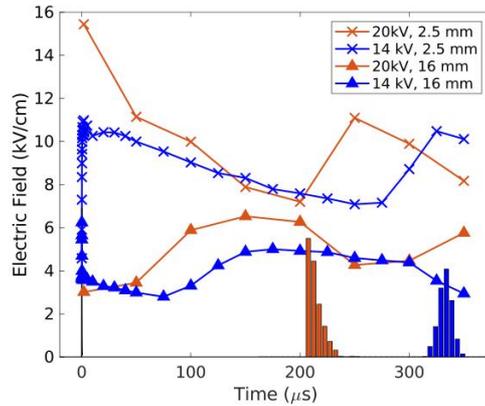
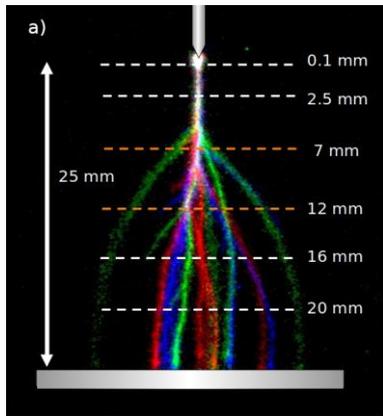


with S. Reuter

[Phys. Rev. Applied 7, 024024 \(2017\)](#)
[Appl. Phys. Lett. 112, 064102 \(2018\)](#)
[Opt. Lett. 44, 3853 \(2019\)](#)

Space-Time resolved E-Field in cold Plasma Jet

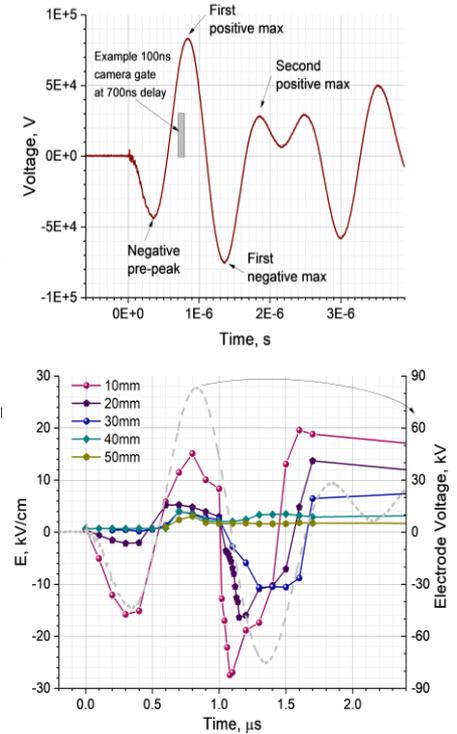
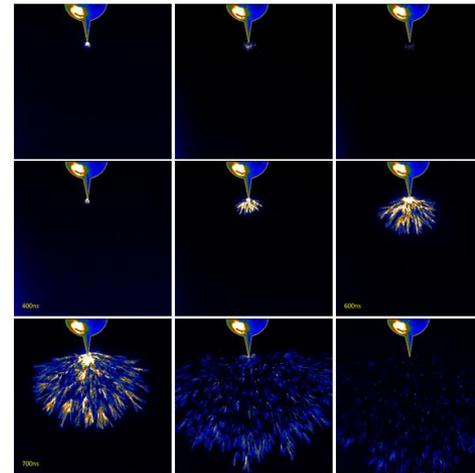
Corona discharge plasma studies using E-FISH



Time-resolved direct measurements of the electric field inside a self-pulsating positive d.c. streamer corona

L. R. Strobel, B. C. Martell, A. Morozov, A. Dogariu, and C. Guerra-Garcia, *Appl. Phys. Lett.*, submitted (2022)

with MIT (PI: Carmen Guerra-Garcia)



Dynamics of volumetric discharge - pulsed streamer corona

S. Elliott, A. Dogariu, T. Cioates, and S. B. Leonov, *Plasma Sources Sci. Technol.*, submitted (2022)

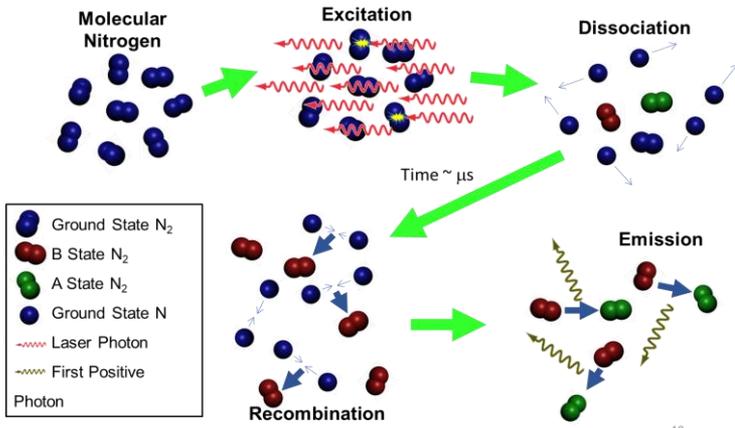
with Notre Dame (PI: Sergey Leonov)

FLEET: Velocimetry using molecular tagging

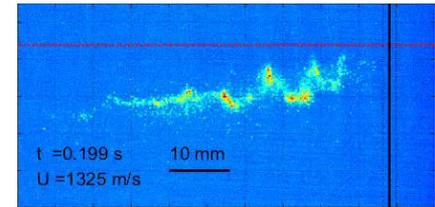
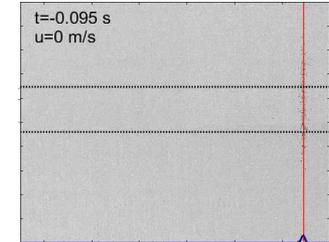
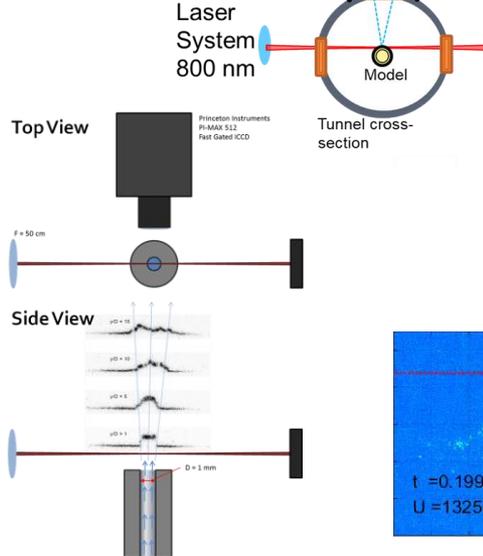
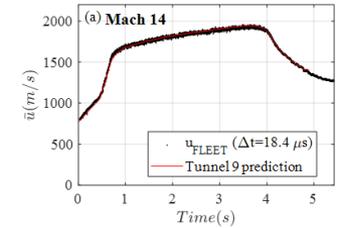
Femtosecond Laser Electronic Excitation Tagging

Uses nitrogen for non-intrusive velocimetry:
Imaging N₂ emission after fs laser dissociation (tagging) and delayed recombination to an excited state

Mechanism for FLEET



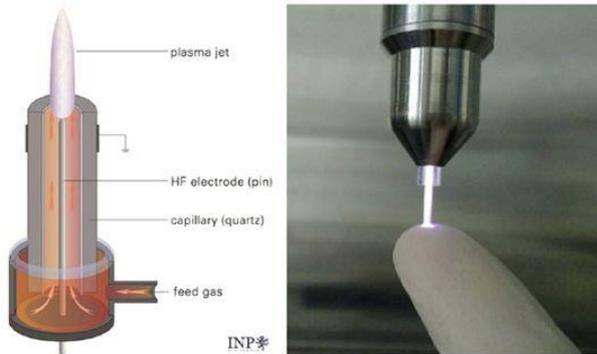
AEDC Tunnel 9



Femtosecond laser electronic excitation tagging for quantitative velocity imaging in air, *Appl. Opt.* **50**, 5158 (2011).
Patent US9863975 (2018)

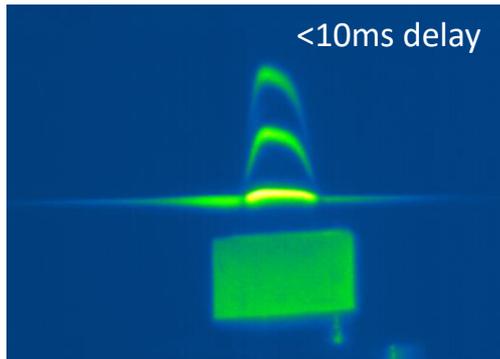
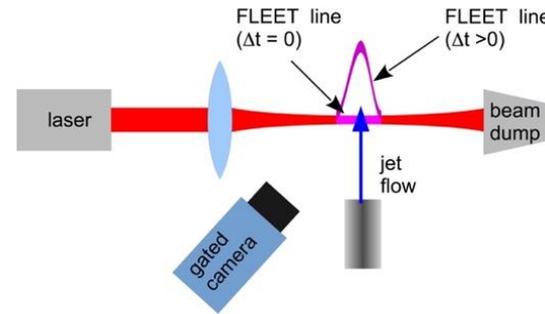
L. E. Dogariu, A. Dogariu, R. B. Miles, M. S. Smith, and E. C. Marineau, "Femtosecond Laser Electronic Excitation Tagging Velocimetry in a Large-Scale Hypersonic Facility," *AIAA Journal* **57**, 4725 (2019).

FLEET: species imaging

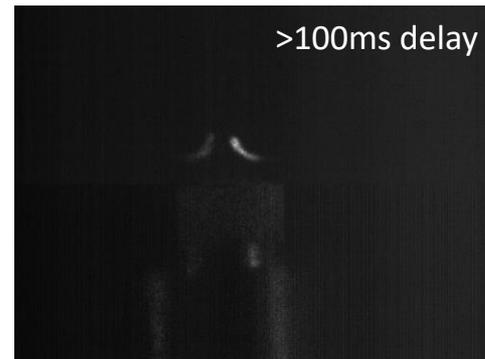


Schematic setup of the APPJ plasma source (left); original plasma jet (right) of the kiNPen © 09.

FLEET in Atmospheric Pressure Plasma Jet (APPJ)

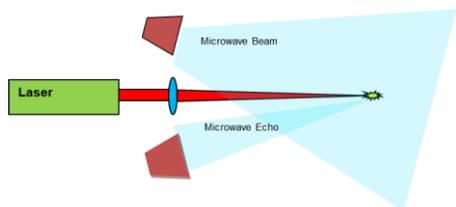


Argon flow velocity mapping

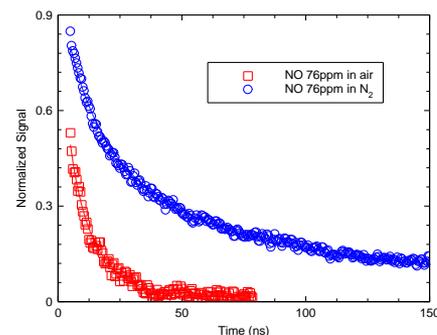
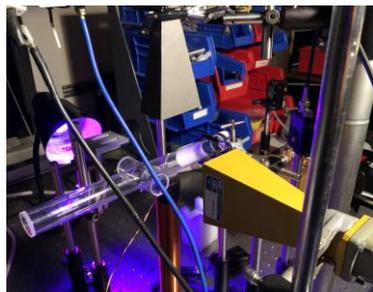


Nitrogen (entrained air)
flow velocity mapping

Radar REMPI (Resonantly Enhanced Multi-Photon Ionization)



- Gas density and temperature, nanoparticle charge, negative ions
- Direct measurement of plasma density and of electron recombination and attachment in air



recombination

attachment

$$\frac{\partial N}{\partial t} = -\nu_a N - \beta N^2$$

$$\beta = 2 \cdot 10^{-13} \sqrt{\frac{300}{T_e(K)}} \frac{m^3}{s}$$

- Homodyne 12-100 GHz system.
- Microwave probes the plasma.
- The mixer output is proportional with the scattering amplitude, hence electron density
- Linear signal from ppm to ppb
- Sub-nanosecond temporal resolution

NO in N_2 - recombination only: $N(t) = \frac{N_0}{1 + \beta N_0 t}$ gives $\beta N_0 = 0.53 \times 10^8 \text{ s}^{-1}$

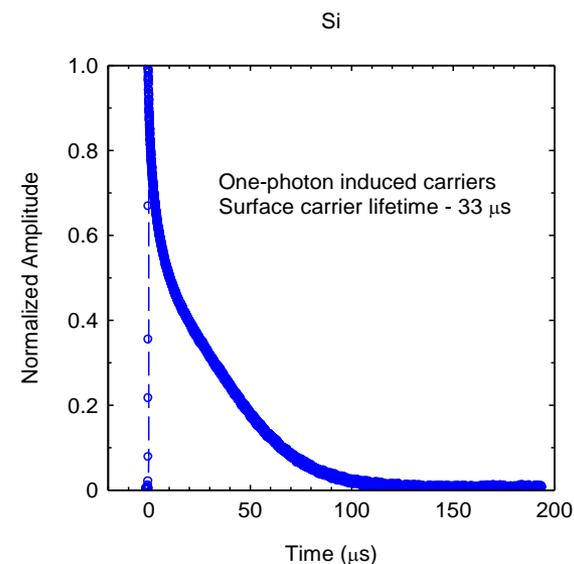
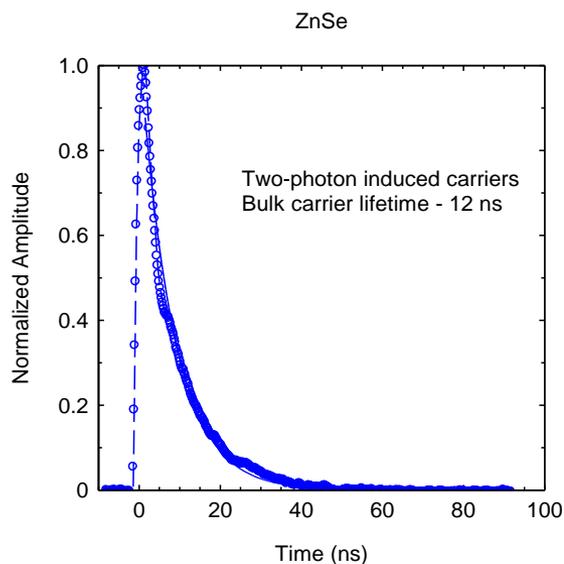
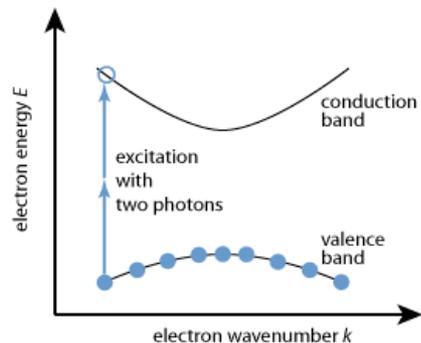
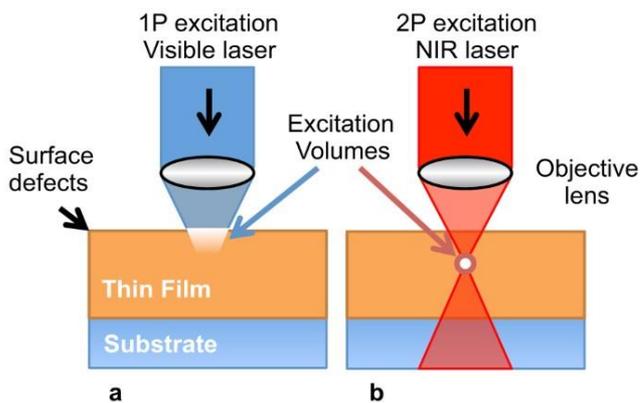
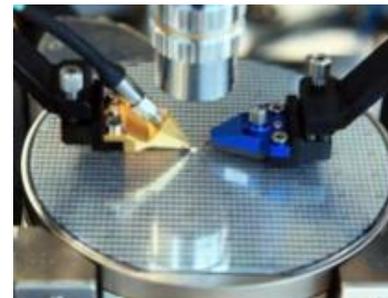
Initial plasma density: $N_0 = 2.5 \times 10^{14} \text{ cm}^{-3}$

NO in air - recombination and attachment: $N(t) = \frac{N_0 e^{-\nu_a t}}{1 + \frac{\beta N_0}{\nu_a} (1 - e^{-\nu_a t})}$
gives $\nu_a = 0.76 \times 10^8 \text{ s}^{-1}$

Dogariu et al, APL 103 224102 (2013)

RADAR REMPI for monitoring electron density on surface and in bulk

- Capability to monitor the doping profile in semiconductors as an *in-situ* alternative to SIMS (Secondary Ion Mass Spectroscopy, *ex-situ*)
- Real-time localized measurements of carrier density, lifetime, bandgap
- Single photon excitation (above bandgap) – surface
- Two photon excitation (below bandgap) – bulk



Patent US7728295 - Method and apparatus for detecting surface and subsurface properties of materials (2010)

(Mobile) Femtosecond Diagnostics in Large Scale Facilities

- **Arnold Engineering Development Complex (AEDC)
Hypervelocity Wind Tunnel 9, White Oak, MD**

First FLEET (velocity) and fs-CARS (temperature) measurements in hypersonic Tunnel 9 (Mach 10,14,18)

A. Dogariu, L.E. Dogariu, M. S. Smith, B. McManamen, J.F. Lafferty R. B. Miles, "Velocity and Temperature Measurements in Mach 18 Nitrogen Flow at Tunnel 9," AIAA SciTech (2021).



- **UT Arlington Aerodynamics Research Center, Arlington, TX**

First FLEET (velocity) and fs-TALIF (species) measurements in hypersonic arc-jet tunnel (Mach 6)

V. Gopal, D. Palmquist, L. Maddalena, L. E. Dogariu, and A. Dogariu, *Exp. Fluids* 62(10), 212 (2021).



- **Princeton Plasma Physics Laboratory**

- Field Reverse Configuration (FRC)
- Hall Thruster Experiment (HTX)

Neutral density (H) in low density plasma ($<10^{10} \text{ cm}^{-3}$)

A. Dogariu, S.A. Cohen, P. Jandovitz, S. Vinoth, E.S. Evans, and C.P.S. Swanson *Rev. Sci. Instrum.*, in print, (2022).

