Two-Dimensional Imaging of Atomic Hydrogen in Pulsed, Non-Equilibrium Nanosecond Plasmas Using Femtosecond TALIF

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• Much work in non-thermal, non-equilibrium plasma discharges generated from nanosecond-duration electrical discharges.
  • Biomedical
  • Materials
  • Combustion
• Concentration measurements of important intermediate species in these non-equilibrium plasmas are still needed.
  • For example, H, N and O in various molecular plasmas.
  • Provide insights into the physical and chemical nature of the plasma system.
  • Facilitate the development of model-based predictive capabilities
Traditional nanosecond two-photon absorption laser-induced fluorescence can be limited by:
- Point-based measurements
- Laser-generated photolytic interferences
- Quenching (must be run at low pressure)

Femtosecond planar two-photon absorption laser-induced fluorescence:
- Enhanced excitation due to larger number of photon pairs from increased bandwidth of near Fourier-transform limited femtosecond pulses.
- Reduced single-photon photo-dissociation due to low average power.

Current work demonstrates proof-of-principle, line imaging of atomic species (H, N) inside low-temperature, nanosecond-pulsed, pin-to-pin, non-equilibrium discharge using fs-TPLIF.
Multi-photon excitation schemes

Two-photon induced fluorescence (TPLIF) Excitation scheme

Other species requiring TP excitation

<table>
<thead>
<tr>
<th>Species</th>
<th>Excitation Wavelength (nm)</th>
<th>LIF Wavelength (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>2 x 205</td>
<td>656</td>
</tr>
<tr>
<td>O</td>
<td>2 x 226</td>
<td>845</td>
</tr>
<tr>
<td>N</td>
<td>2 x 207</td>
<td>745</td>
</tr>
<tr>
<td>Kr</td>
<td>2 x 204</td>
<td>826</td>
</tr>
<tr>
<td>Xe</td>
<td>2 x 224</td>
<td>835</td>
</tr>
<tr>
<td>CO</td>
<td>2 x 230</td>
<td>483</td>
</tr>
</tbody>
</table>

- Collisonal quenching
- Photoionization
- Stimulated emission
- Photolytic production

Experimental apparatus

Laser System
- Regenerative and single-pass ultra-fast amplifier
  - (~1 mJ/pulse at 820 nm)
- Home-built frequency quadrupler
  - (~12 μJ/pulse at 205 nm)

Imaging System
- High-speed visible intensifier, lens coupled to a CCD camera (or CMOS)
- Also used an intensified CCD (ICCD) camera
8-mm diameter electrodes are arranged in a modified pin-to-pin geometry in a 2” diameter, six-way glass cross with UVFS windows.

Top electrode receives +5.0 kV pulsed high voltage from a MOSFET high-voltage switch while the bottom electrode is grounded.

Typical voltage, current and electrical power over an 8-mm gap for 1% hydrogen in helium at 100 torr is shown.
Typical H atom fs-TPLIF Results

Line Imaging

- Externally-intensified (LaVision IRO SA-24) Andor (Newton®) CCD
- 85mm lens with 2” diameter Semrock (BrightLine®) filter
- 16 μm / pixel spatial resolution (with ~30 mm of lens tube extension)
- 300 ns intensifier gate duration; 1 sec CCD exposure; 1000 shots per image
- Typical signal-to-noise ratio of 20:1 (80:1 best case)

Sheet Imaging

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- 1% H₂ in argon at 100 torr
- +5 kV applied to anode
- Collected 25 µs after onset of applied voltage
- 100 mW (avg) laser power
- 22 images used to create reconstructed image
- 75% overlap from image to image
• 1% H₂ in helium at 100 torr
• + 5 kV applied to anode
1D modeling Coupled Poisson equation, Boltzmann equation and non-equilibrium plasma chemistry.

Discrepancies observed between experiment and model could be due to reduced pulse energy observed in model.
Hydrogen Plasma Dissociation

Applied voltage and current plots for:
1% H₂ in He
1% H₂ in Ar

Temporal dissociation plot showing decay of H in 1% H₂ in helium and argon.
Temporal dissociation plot for various mixture fractions in argon at 100 torr.

Temporal dissociation plot for various mixture fractions in helium at 100 torr.

- Production is peaked at 1% admixture in both argon and helium.
- Argon decay rates are very consistent for all admixture percentages.
- Production is 25% higher in helium, but have very similar populations after 50 μs.
- 100% nitrogen at 100 torr
- + 5 kV applied to anode
- Fluorescence is challenging to collect due to strong emission from $\text{N}_2 \text{ B} \rightarrow \text{A (4-2)}$ at $\sim 750\text{nm}$.
- Collected 50 $\mu\text{s}$ after onset of applied voltage
Conclusions

- Development of ultrafast Ti:Sapp laser system
  - Ultrafast amplifier (100 fs FWHM, 10 kHz, 1 mJ/pulse at 820 nm)
  - Home-built quadrupler (~10 μJ/pulse at 205 nm)
  - Enhanced excitation due to larger number of photon pairs from increased bandwidth of near Fourier-transform limited femtosecond pulses.
  - Reduced single-photon photo-dissociation due to low average power.

- Demonstration of 2D imaging in non-equilibrium nanosecond pulsed plasma
  - Obtained temporal decays and 2D reconstruction for H and N atom
  - Temporal delay scans up to 1 ms
  - Mixture fractions of 1% and 5% of H₂ in both He and Ar background gasses
  - Kr calibration for quantitative H atom populations
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Fs-duration Ti-Sapp laser with regen cavity and single-pass amplifier system produces ~100 fs pulses at 800 nm with 20 nm bandwidth.

Fundamental is mixed with third harmonic in a home-built frequency quadrupling unit to generate UV radiation consisting of 10 kHz, 12 µJ pulses near 200 nm.

Schemes to pump atomic species through two-photon absorption in UV\textsuperscript{1,2} are shown at right.

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Intense laser pulses can photochemically produce atomic hydrogen

H-atom LIF line profile (ns excitation)

Main photolytic precursors for H:
- $\text{H}_2\text{O}$ ($\text{H}_2\text{O} + \text{hv} \rightarrow \text{H} + \text{OH}$)
- $\text{CH}_3$ ($\text{CH}_3 + \text{hv} \rightarrow \text{CH}_2 + \text{H}$
  - $\text{CH}_2 + \text{hv} \rightarrow \text{CH} + \text{H}$
  - $\text{CH} + \text{hv} \rightarrow \text{C} + \text{H}$)

Short pulse excitation can enable interference-free detection via TPLIF

- Photolytic production scales linearly with pulse energy for single-photon photo-dissociation:
  \[ N_{H_{ph}} \propto \int I dt \sim \left( \frac{E_L}{\Delta t} \right) \Delta t = E_L \]

- TPLIF signal scales quadratically with intensity:
  \[ S_{LIF} \propto \int I^2 dt \sim \frac{E_L^2}{\Delta t} \]

- Short-pulse excitation reduces pulse energy required for two-photon excitation
  - Can it reduce interference?

Interference-free line imaging of H atoms in methane flames

ns excitation
(~8 ns)

- Radial Position (mm)
- Normalized LIF Signal
- 10-20 Hz, Shot-averaged


Premixed, CH₄/O₂/N₂ flame

ps excitation
(~100 ps)

- Radial Position (mm)
- Normalized LIF Signal
- 10 kHz, Single-shot


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