Two dimensional laser-collision induced fluorescence measurements in low pressure plasmas

AVS 58th Annual International Symposium
Nashville, TN November 3'rd 2011

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This work was supported by the Department of Energy Office of Fusion Energy Science
Contract DE-SC0001939
Laser-collision induced fluorescence provides measure of electron density and "temperature"

**Motivation:** What is the density? What is the temperature? Where and When?
- More traditional probe techniques may couple and perturb
- Optically passive techniques are line-of-sight limited
- Optically active-techniques such as Thomson scattering pose their own set of challenges

**In this presentation**
- Laser-collision induced fluorescence (LCIF) primer
  - Collisional-radiative model used to predict LCIF
  - Applied to triplet manifold of Helium
- Implement and benchmark technique
  - Experimental setup
  - Time evolution of LCIF and time integrated LCIF
- Applications of LCIF:
  - Ion sheaths, transient double layers, positive columns
- Future directions and concluding comments
  - Extension of helium
  - Investigate argon
Part I: LCIF concepts and key trends

- **Overview**
  - LCIF concepts
  - Collisional-radiative model for helium
  - Key scaling trends

**LCIF Concept**

- Electron collisions
- Laser excitation
- \( \Delta N_i \)

**CR Model**

\[
\frac{dN_j}{dt} = \left[ \sum_{i \neq j} K_{ij}^e N_i - \sum_{i \neq j} K_{ji}^e N_j \right] n_e + \ldots.
\]

**Key scaling trends**

- Ratio to 389 nm
- Electron density (cm\(^{-3}\))
- Electron temperature, \( T_e \) (eV)
LCIF is based on redistribution of excited state by plasma electrons

- Laser excitation causes populates an intermediate state
  - Relaxation processes deplete excited state
- Portion of excited state population gets redistributed into "uphill" states
  - Driven by interaction with energetic plasma electrons

$\Delta N_j \sim K_{ij}^e n_e \times \Delta N_i \times \Delta t$

Electrons coupled interaction

Change in upper state due to excited state

$\text{Photons emitted} \sim A_{jk} \times \Delta N_j \times \Delta t$

Detected light

LCIF looks for changes in emission of neighboring states after laser excitation
Redistribution after laser excitation is complex

- A "good" model is required to predict transfer between levels
  - Employ a collisional-radiative model (CRM) to predict redistribution

\[
\frac{dN_j}{dt} = \left( \sum_{i \neq j} K_{ij}^e N_i - \sum_{i \neq j} K_{ji}^e N_j \right) n_e + \sum_{i > j} A_{ij} N_i - \sum_{i < j} A_{ji}^j N_j + \sum_k \left[ \sum_{i \neq j} K_{ikj}^a N_i - \sum_{i \neq j} K_{jki}^a N_j \right] N_k
\]

- Electron density and electron temperature appear in first term
  - Temperature dependence introduced via \( K_{ij}^e \)

\[
K_{ij}^e = \left\langle \sigma_{ij} (E) v_e \right\rangle
\]

\[
f(v) \sim e^{-\frac{(1/2mv)^x}{kT}}
\]

Approach is applicable to various atomic and molecular systems of interest
Helium atom serves as target species for LCIF measurements

- Employ Helium to start with - considering argon
  - "Simple system" with "better known" rates
- Utilize functionalized form of cross-sections compiled by Ralchenko
  - Integrate to get rates, compare to measured rates

Key transitions

<table>
<thead>
<tr>
<th>Key transitions</th>
<th>Computed and measured excitation rates in Helium</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^3S \rightarrow ^3P$, $^3D$</td>
<td><img src="image1" alt="Graph 1" /></td>
</tr>
<tr>
<td>$^3P \rightarrow ^3S$, $^3D$</td>
<td><img src="image2" alt="Graph 2" /></td>
</tr>
<tr>
<td>$^3P \rightarrow ^4S$, $^4P$, $^4D$</td>
<td><img src="image3" alt="Graph 3" /></td>
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</tbody>
</table>


**Accuracy of $n_e$, $T_e$ depend on knowledge of $K_{ij}(kT_e)$**
CRM predicts evolution of various helium states after laser excitation

- Temporal evolution serves as a partial "fingerprint" of electron interaction
  - Analyze shape of decay above \( n_e \sim 10^{11} \) electrons/cm\(^3\)
  - Below \( n_e \sim 10^{11} \) absolute intensities are needed

\[
K_{ij}^e n_e \sim A_{ij} \quad 10^{-5} \times 10^{11} ~ 10^7
\]

Computed temporal evolution

Key transitions

\[
\begin{align*}
3^3P &\rightarrow 2^3S \\
3^3S &\rightarrow 2^3P \\
3^3D &\rightarrow 2^3P \\
4^3D &\rightarrow 2^3P
\end{align*}
\]

Normalized LCIF

Time (ns)

\( n_e = 10^{10} \) e/cm\(^3\)
\( n_e = 10^{11} \) e/cm\(^3\)
\( n_e = 10^{12} \) e/cm\(^3\)

\( kT_e = 2 \) eV

Need at least two time-resolved profiles to uniquely obtain \( n_e, kT_e \)
Time integrated intensity trends are utilized instead of time resolved LCIF

- Examine ratios of time integrated LCIF
  - Eliminates need for absolute calibrations
  - Still need relative efficiencies of imaging system
- Capitalize on "$kT_e$ independent" coupling of $3^3P$ to $3^3D$
  - Ratio of 588 nm to 389 nm yields $n_e$
  - Density + Ratio of 447nm to 588 nm yields $kT_e$

\[
\frac{\Delta N_j}{N_{3^3P}} \sim K^{e}e_{3^3P \rightarrow j} n_e \times \Delta t
\]

\[
\frac{\Delta N_j}{\Delta N_k} \sim \frac{K^e_{0j}}{K^e_{0k}}
\]

Only need to make three measurements to obtain $n_e$, $kT_e$
Small energy gap leads to "$kT_e$ independent" coupling of $3^3P$ to $3^3D$

Considerable fraction of the electrons are capable of driving the interaction

Part II: LCIF implementation and benchmark

- Implement and benchmark technique
  - Experimental considerations
  - Benchmarking LCIF - compare observations with anticipated trends

**Experimental setup**

![Diagram of experimental setup]

**Benchmark LCIF**

![Graphs showing normalized LCIF vs. time (ns)]

**Density Trends**

Electron densities (e/cm$^3$)

<table>
<thead>
<tr>
<th>Set</th>
<th>CR predictions</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td></td>
</tr>
<tr>
<td>B</td>
<td></td>
</tr>
<tr>
<td>C</td>
<td></td>
</tr>
</tbody>
</table>

588/389 ratio

- 0.1
- 1

Data Set A: $A_{Eff} = A_{Nom}$

Data Set B: $A_{Eff} \gg A_{Nom}$ (During laser excitation)

**Other components**

- Delay generator
- Pulsed laser
- Beam Expander
- Gated ICCD Camera
- V$\text{Mod}$
- Gas in
- Gas out
- 80 GHz
- Filtered PMT
- To digitizing oscilloscopes
- Pulse + DC voltage
Experimental implementation of the LCIF is realized

- Nanosecond pulsed laser used for excitation
  - < 10 ns FWHM, < 0.1 cm\(^{-1}\) line width
- Timing of experiment controlled by delay generators
  - Move experiment and imaging with respect to firing of the laser
- Image LCIF with gated-intensified CCD
  - Narrow (~ 1 nm FWHM) interference filters centered on lines of interest
- Take two images per transition considered
  - Total emission and plasma induced emission (PIE) - subtract the two

**Optical setup**

**Timing sequence**

Need to make six (3 x 2) measurements to obtain \(n_e, kT_e\)
Pulsed positive column is utilized to benchmark LCIF technique

- Pulse discharge currents generate broad density range
  - ~ 10 Microseconds, 80 GHz interferometer
- Compute drift velocities and extract electron temperatures
  - Use published drift parameters

Positive column


Positive column is a good vehicle to benchmark LCIF technique

Helium drift parameters

E/N (Td) vs. Electron Temperatures (eV)

Drift velocity (cm/s)
First steps: Verify time resolved LCIF to test CRM

- First sets of measurements presented some surprises
  - Strong radiative coupling between $3^3P$ and $3^3S$ states
- Proper accounting produced observed trends
  - Measured data and predicted behavior are consistent

Key transitions

```
\begin{align*}
3^3P & \rightarrow 2^3S \\
& \quad 389 \text{ nm} \\
3^3S & \rightarrow 2^3P \\
& \quad 707 \text{ nm} \\
3^3D & \rightarrow 2^3P \\
& \quad 588 \text{ nm} \\
4^3D & \rightarrow 2^3P \\
& \quad 447 \text{ nm}
\end{align*}
```

Representative results

- Red: CRM predictions ($n_e = 5 \times 10^{10}$, $T_e \sim 4 \text{ eV}$)
- Blue: Measured LIF/LCIF

Good first demonstration of LCIF technique
[588]/[389] ratio exhibits linearity over nearly two orders of magnitude

- Better yet, measured ratios agree reasonably well with computed ratios
  - Slightly higher, and some deviation at low density
- Examined trends at different times during the current pulse
  - Anticipate different temperatures as column is established

Waveforms during excitation

Density dependent ratio trends

Density measurements obtained at different times essentially overlay each other
[447]/[588] ratio captures trends but misses absolutes

- Anticipated $T_e$ trends are observed
  - High temperature at start, low temperatures later on
- Measure $T_e$ trends mimic computed trends
  - Discrepancy in absolute values are apparent

Uncertainties in rates, EEDF and/or interpolation of $T_e$ from drift parameters should impact absolute values
Part III: Applications of LCIF

- Applications of LCIF
  - ion sheaths, double layers and positive columns

**Emphasize structure and evolution of the plasma being studied**
Demonstration of LCIF technique: 2D-sheath formation

- Examine evolution and structure of ion sheath
  - 1 kV bias applied to inner electrode, 50 μs into afterglow (low n_e, low kT_e)
  - 20 ns snapshots of LCIF, 30 ns steps

Data

Time = -30 ns

Time = +0 ns

Time = +30 ns

Time = +70 ns

Time = +2000 ns

Setup

Electron Density (10^9 e/cm^3)

Decent temporal and spatial resolution demonstrated

Initial structure

Voltage applied

"Matrix" sheath

Sheath evolving

Final sheath structure

Demonstration of LCIF technique: 2D-sheath formation

- Examine evolution and structure of ion sheath
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Setup

Electron Density (10^9 e/cm^3)

Decent temporal and spatial resolution demonstrated

Initial structure

Voltage applied

"Matrix" sheath

Sheath evolving

Final sheath structure
Interesting structure observed in the sheath

- LCIF signal observed deep in the sheath
  - Some caused by neutrals, but not all

**2D Images**

- "on axis"
- "off axis"

**Profiles**

Signal deep in the sheath caused by electrons emitted from the electrode
Electrons backfill ion sheath after voltage is removed

- LCIF detects electrons but not ions
  - Examine time immediately after voltage is removed

Ion densities can be quantified after voltage is removed.
Double layer more pronounced in ECR based plasma cathodes

- NASA driven research interested in electron sources for propulsion
  - Understand limitations on current extraction
- Host Brandon Weatherford (U. Mich.) to implement LCIF
  - Examine coupling of between plasma generation and electron extraction

Multi-structure plasma formed by electron-extracting electrode...

... quite difficult to probe with more conventional means!
LCIF is being used to study structure in a positive column

- Positive column is "well understood" system
  - Studied extensively, use it for calibration
- Platform for fast ionization wave (FIW) studies
  - Observations may warrant their own study

Benchmark 2D simulations with measurements made by LCIF

Part IV: Future pursuits with LCIF

- Extension of LCIF technique to other operating regimes
  - Limitations of helium and paths around this….

**Extension of helium**

- $2^3S @ 10$ Torr
- $2^3P @ 10$ Torr
- $2^3P @ 100$ mTorr

**Argon LCIF**

- "Orange" LCIF from 4d states well separated from "red" lines emanating from 2p states
- Targeting lowest lying $3p_{10}$ state
- LIF at 470.2 is well isolated from other transitions
- Coupled to $1s_2$ state
- Not pursuing radiation trapped 2p states

Electron Density (cm$^{-3}$)

Excited State Density (cm$^{-3}$)
Helium becomes limited at higher pressures

- **Helium proved to be well suited for lower pressure and lower densities**
  - Limited spectroscopic pathways
  - Well known cross-sections
  - Highly populated, long lived $2^3S$ metastable state

- **As density increases and as composition changes**
  - Radiation trapping/transport becomes problematic

\[
\tau = \frac{L}{l_{mf}} \approx f_{nm} \lambda \left( \frac{Mc^2}{kT_A} \right)^{1/2} N_A L \ll 1
\]

<table>
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<td>$3^3P \rightarrow 2^3S$</td>
<td>0.064</td>
<td>&gt;1</td>
</tr>
<tr>
<td>$4^3P \rightarrow 2^3S$</td>
<td>0.02</td>
<td>~1</td>
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(Assuming $L=1$ cm and $N_A \sim 10^{13}$ absorbers/cm$^3$)

*States connected to $2^3S$ can become trapped due to strong coupling and higher populations*
Pump out of alternative helium states

- At higher pressures and densities, $^2\!^3P$ state becomes adequately populated
  - Comparable oscillator strengths (into comparable levels)
  - Sufficiently lower population compared to $^2\!^3S$

Scaling

$^2\!^3S$ State:

$$n_{^2\!^3S} \sim \frac{K_{0 \rightarrow S}^e}{K_{S \rightarrow P}^e} n_0 \sim 10^{-5} n_0$$

$^2\!^3P$ State:

$$n_{^2\!^3P} \sim \frac{1}{A_{P \rightarrow S}} \left[ K_{0 \rightarrow P}^e + K_{S \rightarrow P}^e \right] n_0 n_e$$

Trends

Low $^2\!^3P$ state densities should be high enough to pump but low enough not to trap
Spectroscopic pathway proposed for pumping from $2^3P$

- Lower base density of $2^3P$ advantageous, but some tradeoffs
  - Lose the nice "temperature free" $3^3P \rightarrow 3^3D$ transition
  - Spectrally dense - many transitions ~ 400 nm

Previous approach

<table>
<thead>
<tr>
<th>State</th>
<th>Energy Difference</th>
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<tr>
<td>$2^3S$</td>
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<td>$3^3P$</td>
<td>$\Delta E \sim 0.07$ eV</td>
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<td>$\Delta E \sim 0.7$ eV</td>
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<td></td>
</tr>
<tr>
<td>$5^3S$</td>
<td>$\Delta E \sim 0.38$ eV</td>
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Proposed approach

<table>
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<td>$2^3S$</td>
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<tr>
<td>$3^3S$</td>
<td>$\Delta E \sim 0.14$ eV</td>
</tr>
<tr>
<td>$3^3D$</td>
<td></td>
</tr>
<tr>
<td>$4^3D$</td>
<td></td>
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Pumping to the $3^3S$ or the $3^3D$ states are not "off the table" and may be perused
Preliminary investigation of proposed scheme looks promising

- Employ same pulsed positive column used for $2^3S$ excitation
  - Limit observations to states coupled to $2^3P$
  - Integration of 20 ns, 10 ns after laser excitation
Preliminary investigation of proposed scheme looks promising.
Alternative gasses are being considered

- **Technique is extendable to other gases**
  - Helium is seldom "gas of choice"
  - Helium becomes problematic in mixtures

- **Argon is commonly used gas and obvious choice**
  - More pump-probe pathways to consider
  - Individual lower lying $1s_x$ states are anticipated to less populated

\[ \tau = \frac{L}{l_{mfp}} \approx f_{nm} \lambda \left( \frac{Mc^2}{kT_A} \right)^{1/2} N_A L << 1 \]

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<tr>
<td>$2p_{10} \rightarrow 1s_5$</td>
<td>0.17</td>
<td>&gt;&gt;1</td>
</tr>
<tr>
<td>$3p_{10} \rightarrow 1s_5$</td>
<td>$9 \times 10^{-4}$</td>
<td>0.08</td>
</tr>
</tbody>
</table>

(Assuming $L=1$ cm and $N_A \sim 10^{13}$ absorbers/cm$^3$)

*Low oscillator strengths and possibly lower densities make argon attractive*
Complexity of argon makes extension of LCIF "challenging"

- Argon offers more spectroscopic pathways to pursue

Targeting lowest lying $3p_{10}$ state

Not pursuing radiation trapped $2p$ states

LIF at 470.2 is well isolated from other transitions Coupled to $1s_2$ state

"Orange" LCIF from 4d states well separated from "red" lines emanating from 2p states

Cross sections and rates not well known for electronic driven processes from 3p to higher states

Taken from Bogearts et. al, J. Appl. Phys. 84, 121, 1998
Despite reservations Argon LCIF is being investigated

- Well characterized positive column is used to test feasibility
  - Microwave interferometer to measure densities
  - Pulsed for higher densities/temperatures and stable plasmas
  - Time resolved LIF/LCIF with PMT + narrowband filters

**Calibration cell**

**Preliminary trends**

**Perspective transitions are identified and calibration of the technique is underway …. Stay tuned!**
Concluding remarks and future directions

- **LCIF technique demonstrated in 2D**
  - Free of “line of sight” constraints
  - Good spatial resolution – limited by optical collection
  - Decent temporal resolution – limited by ICCD gate times & tolerable signals

- **Caution required for proper implementation of the technique**
  - Uncertainties about rates – Absolute bounds on measurements
  - Proper choice of model – Capture the required physics

- **Technique should be extendable over broad parameter space**
  - Higher pressures – neutral collisions
  - Smaller dimensions – scattering and access
  - Other atomic systems

*This work was supported by the Department of Energy Office of Fusion Energy Science
Contract DE-SC0001939*
Thank you
Neutral mixing of $3^3P$ and $3^3D$ needs to be considered

- Proximity (energetically) of states means neutrals can transfer excited population to $3^3D$
  - Energy spacing between states is $0.067 \text{ eV} \sim 780 \text{ K}$

Amount of $3^3D$ produced from $3^3P$

$$\Delta N_{Electrons} \sim K_{P\rightarrow D}^e n_e \Delta N_P \Delta t$$

and

$$\Delta N_{Neutrals} \sim K_{P\rightarrow D}^N n_0 \Delta N_P \Delta t$$

Bound determined by setting the two equal

$$\frac{\Delta N_{Electrons}}{\Delta N_{Neutrals}} \sim \frac{K_{P\rightarrow D}^e n_e}{K_{P\rightarrow D}^N n_0} \sim 1$$

Solve for $n_e$

$$n_e \sim \frac{K_{P\rightarrow D}^N n_0}{K_{P\rightarrow D}^e} \sim \frac{10^{-12}}{10^{-5}} n_0$$

Lower limit on electron density scales with pressure (and temperature) of neutral background

References for rates and cross-sections

- **Superelastic**
  - Klein Rosseland
  - Sobelman

\[ K^e_{ij} = \langle \sigma_{ij} v_e \rangle = \left( \frac{m_e}{2 \pi k T_e} \right)^{\frac{3}{2}} \int_0^\infty \sigma_{ij}(v) \exp \left( \frac{-m_e v^2}{2 k_B T_e} \right) 4 \pi v^3 \, dv \]

Key uphill transitions are not significantly impacted by radioactive coupling

- Dominant population pathway is still through excited $3^3P$ state
  - Final densities of $3^3P$ state will change, but this is normalized out in analysis

Averaged trends

Data Set A: $A_{Eff} = A_{Nom}$

Data Set B: $A_{Eff} >> A_{Nom}$ (During laser excitation)

Ratio to $\lambda = 389$ nm

Electron density (cm$^{-3}$)

$\lambda = 389$ nm
$\lambda = 707$ nm
$\lambda = 588$ nm
$\lambda = 447$ nm

$kT_e = 2$ eV

$A_{Eff} = A_{Nom}$

$A_{Eff} >> A_{Nom}$

Caution should still be used if considering the $3^3S$ state for normalization
Analysis of ion-matrix sheath is used to further test LCIF technique

- Immediately after the pulse is applied
  - Electrons are rapidly pushed away from electrode
  - But ions should have little time to respond

- Using Poisson’s equation and assuming quasi neutrality before the application of the pulse:
  \[ \nabla^2 V = \frac{e}{\varepsilon_0} n \rightarrow n_i \approx \frac{2\varepsilon_0 V}{e\Delta x^2} \]

- For a voltage of 1 kV and a thickness of ~ 5 mm:
  \[ n_e \sim 4 \times 10^9 \text{ electrons/cm}^3 \]

*Reasonable cross-check of LCIF technique*