

Ultrafast Laser Diagnostics to Interrogate High Pressure, Highly Collisional Plasma Environments

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Part I: OVERVIEW AND EXTENSION OF LCIF TO HIGHER PRESSURES

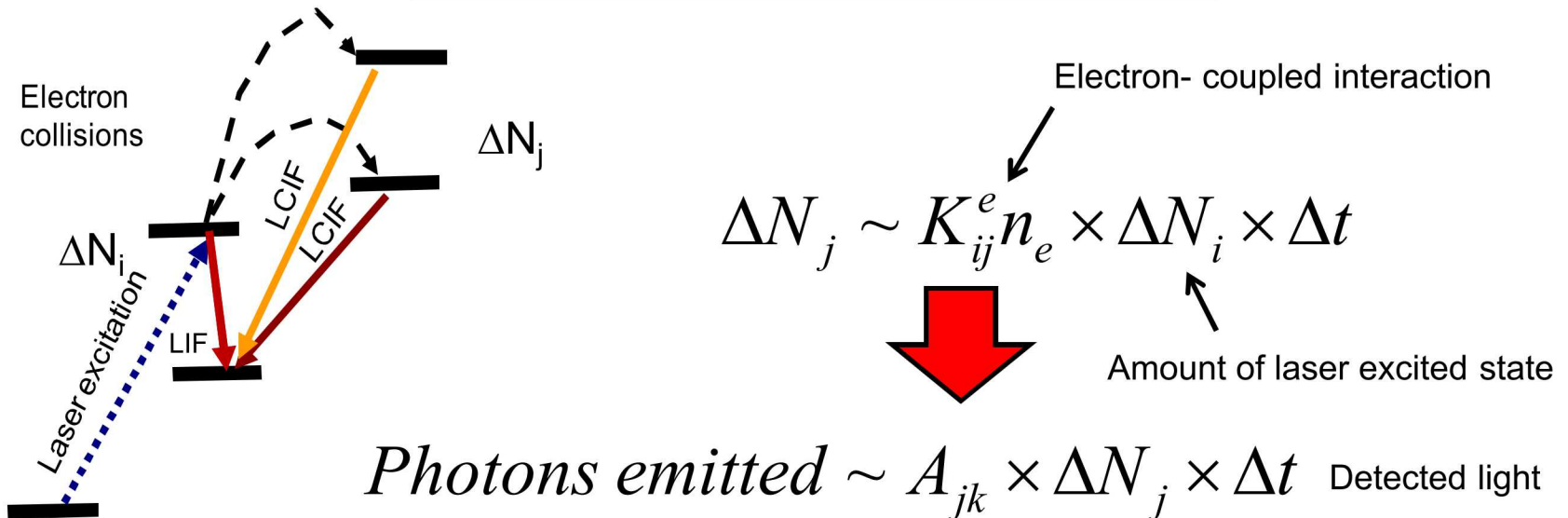
- **Part I: Overview of laser-collision induced fluorescence (LCIF)**
 - **LCIF Concepts and challenges with LCIF at higher pressures**
 - **Extension of LCIF and demonstration of key LCIF trends**
- **Part II: Application to various discharge systems of interest**
 - **Interrogate structured and dynamic systems**

Discuss concepts and demonstrate key scaling trends under relevant conditions

SIMPLIFIED OVERVIEW OF LCIF

- LCIF depends on collisionally induced redistribution of a laser excited state into near by states
 - Appropriately interactions can populate energetically “uphill” states.

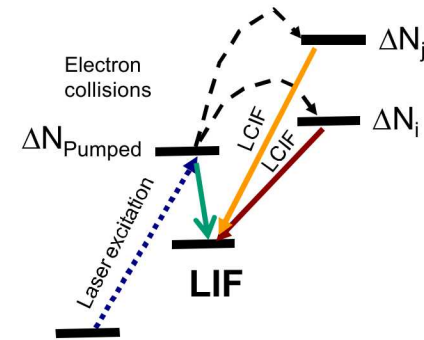
Laser Collision Induced Fluorescence



LCIF signals can depend on electron density (n_e) and E/N (K_{ij}^e)

CHALLENGES OF EXTENDING LCIF TO HIGH PRESSURE

- Observed LCIF is superposition of several complex processes.
 - In general need a good model to describe the redistribution.



$$\frac{dN_j}{dt} = \underbrace{\left[\sum_{i \neq j} K_{ij}^e N_i - \sum_{i \neq j} K_{ji}^e N_j \right] n_e}_{\text{"Electron mixing"}} + \underbrace{\sum_k \left[\sum_{i \neq j} K_{ikj}^a N_i - \sum_{i \neq j} K_{jki}^a N_j \right] N_k}_{\text{"Neutral mixing"}} + \underbrace{\left[\sum_{i > j} A_{ij} N_i - \sum_{i < j} A_{ji}^j N_j \right]}_{\text{"Photon mixing"}}$$

Low pressure approximation

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

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

Simplifications likely not to be so forthcoming at higher pressures

EXTENDING LCIF TO HIGH PRESSURE PRESENTED CHALLENGES

- Several challenges need to be considered at higher pressures
 - Heavy-particle interactions place lower bound on electron detection.
 - Collisions reduce lifetime of laser excited states.
 - Line broadening can limit access to interrogated state.

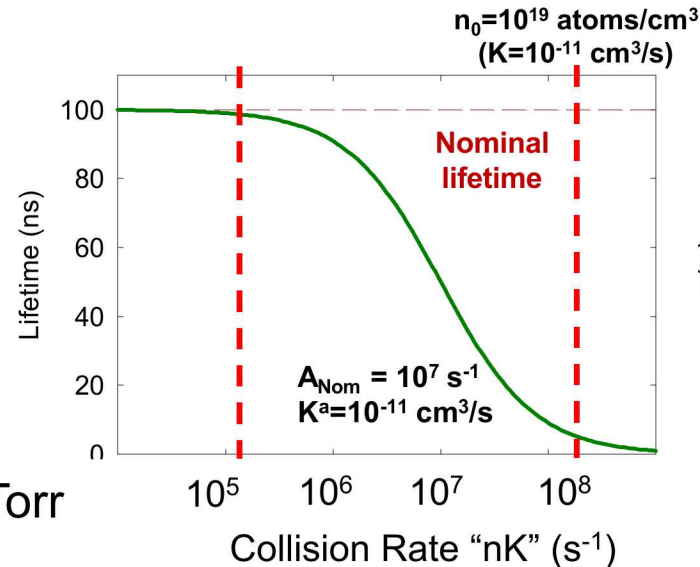
Bounds on detection

$$\frac{\Delta N_{\text{Electrons}}}{\Delta N_{\text{Neutrals}}} \sim \frac{K^e n_e}{K^N n_0} \sim 1$$

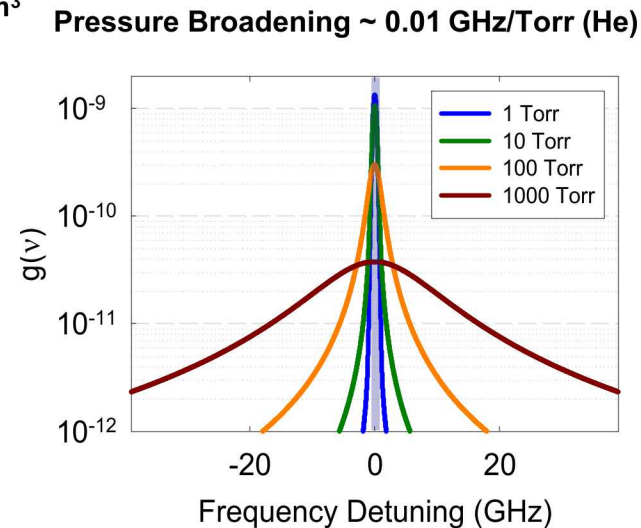
$$n_e \sim \frac{K^N}{K^e} n_0 \sim \frac{10^{-11}}{10^{-5}} n_0$$

Limit $\sim 10^{13}$ e/cm³ at 1000 Torr

Reduced lifetimes



Absorption profiles

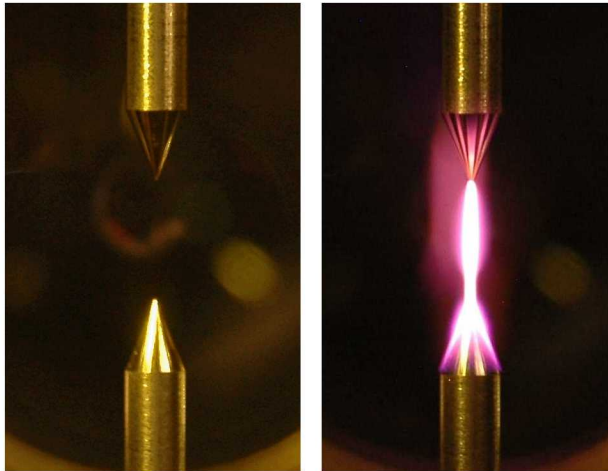


Short pulse lasers coupled with fast gated imaging helped face challenges.

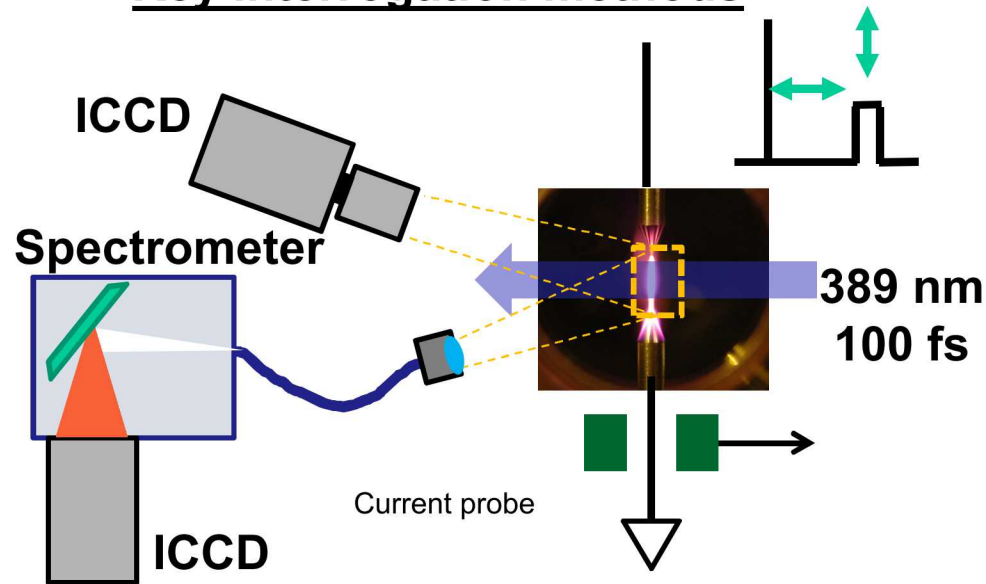
KEY CHALLENGE: GENERATING AND MANIPULATING WELL CHARACTERIZED PLASMA

- Need plasma with well controlled n_e , E/N to calibrate LCIF.
 - Plasma generation in 640 Torr He.
 - Double pulse method to separate generation and interrogation.
 - Spectrometer to identify, camera to image.

Discharge configuration



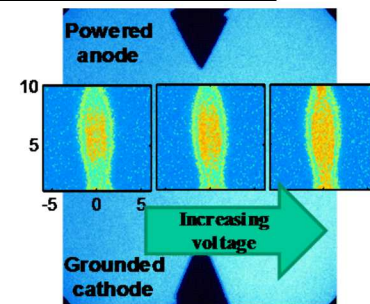
Key interrogation methods



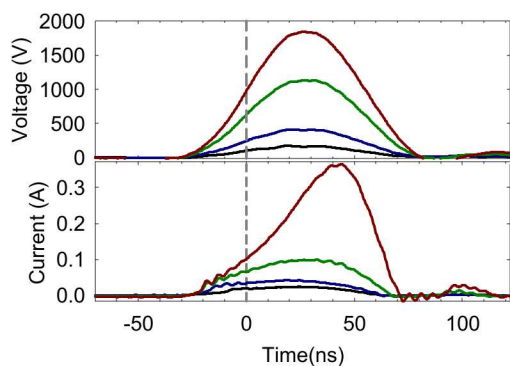
Setup enables good access and control of the plasma for LCIF calibration.

DOUBLE PULSE PLASMA SEPARATES GENERATION FROM MANIPULATION

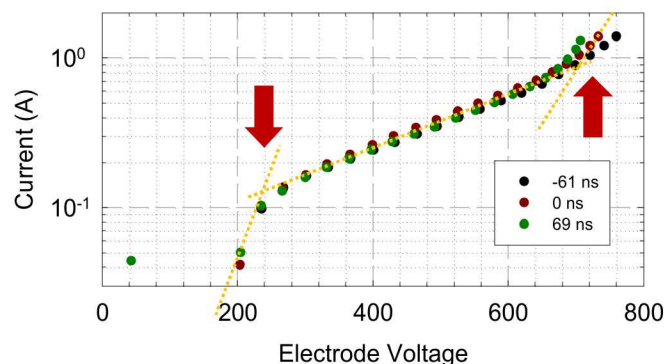
- Need plasma with well controlled n_e , E/N to calibrate LCIF.
 - First voltage pulse dictates plasma density and distribution.
 - Second pulse drives current through afterglow.



Sample Response



Extracted Current



Anticipated plasma parameters

$$n_e = \frac{m_e v_m N I}{e^2 N E A}$$

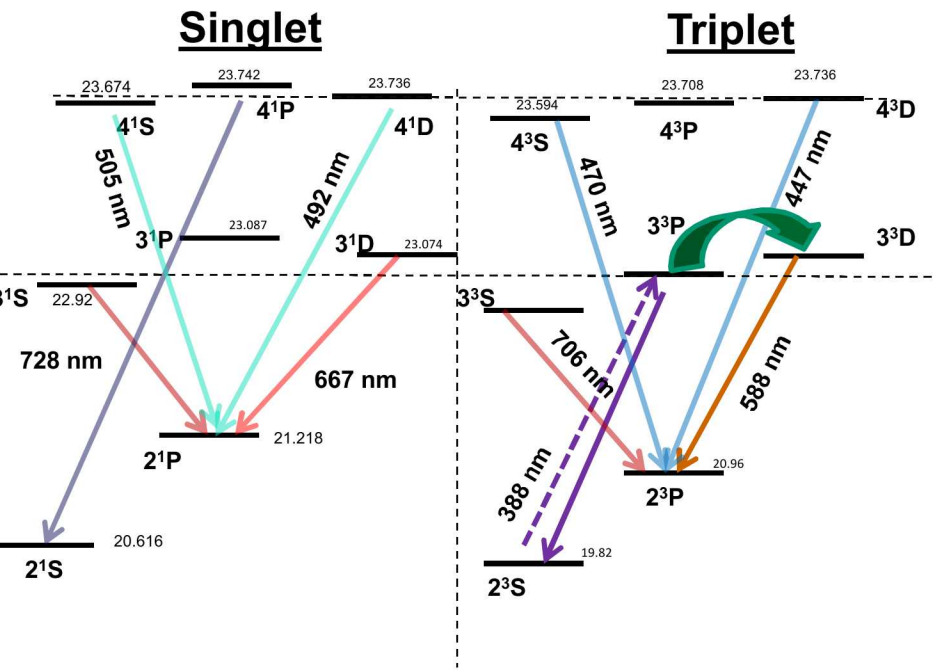
V	I	J	E/N	Vd	n_e
V	A	A/cm ²	Td	cm/s	e/cm ³
225	0.1	3.18	0.1	1.50E+05	1.32E+14
700	0.6	19.10	5	1.00E+06	1.19E+14

Structure of the filament does not change significantly as it is “heated” – at least initially.

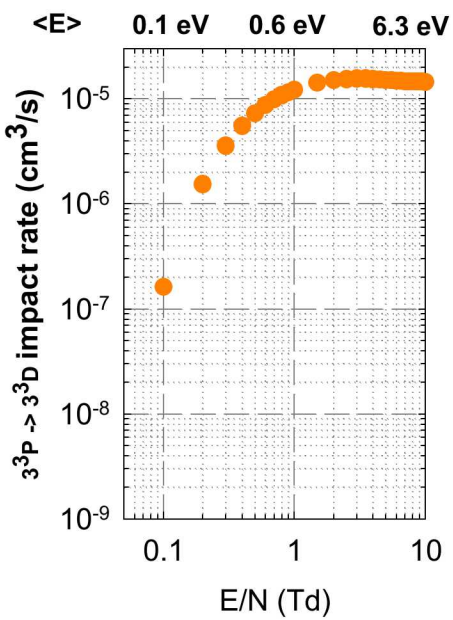
RATIO OF LCIF LINES ARE UTILIZED TO IDENTIFY KEY SCALING TRENDS

- Coupling into 3^3D LCIF is work-horse for electron density measurements.
 - At higher E/N scaling becomes independent of E/N.

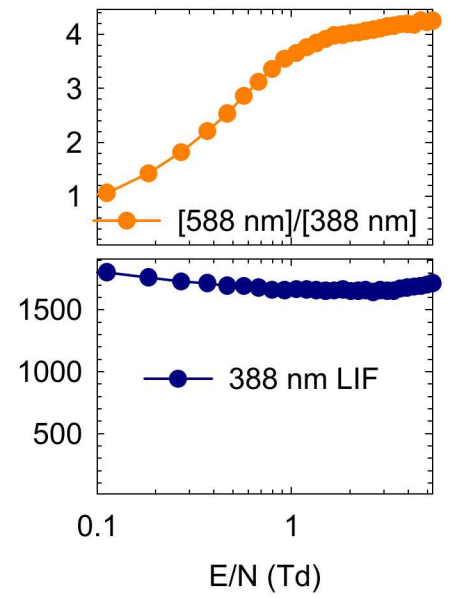
Pathway



Rates



Scaling of LCIF



3^3D LCIF provides strong signals over broad range of plasma conditions.... but demonstrates E/N dependence below 1 Td.

RATIO OF LCIF LINES ARE UTILIZED TO IDENTIFY SCALING TRENDS

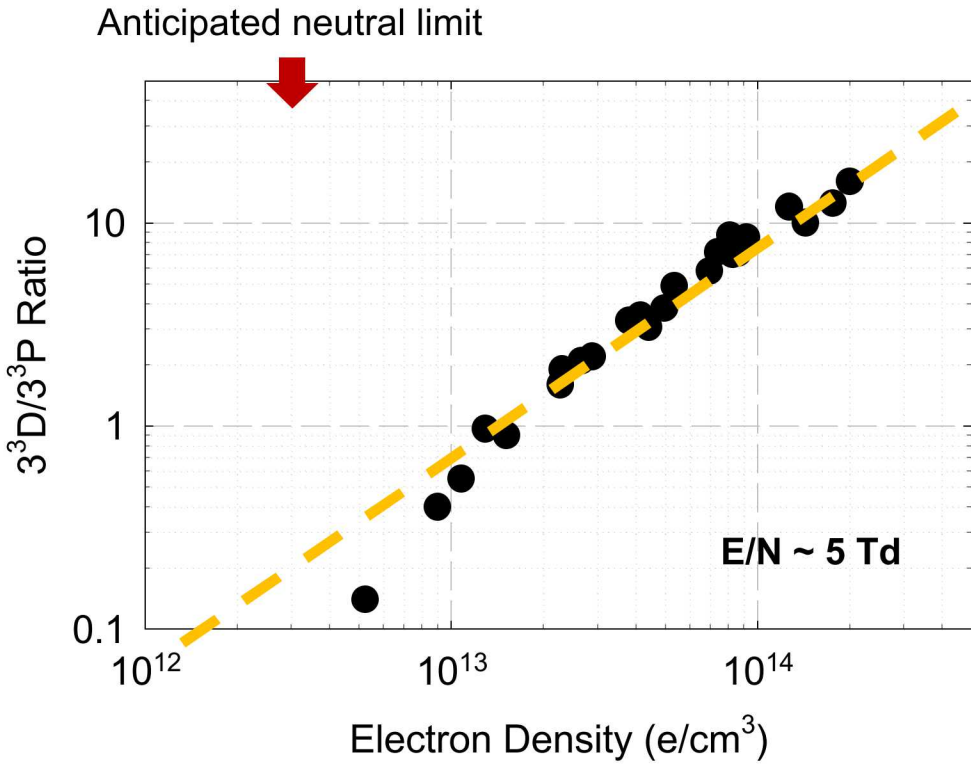
- Benchmark scaling of density dependence of 3³D LCIF.
 - Utilize the E/N invariance of 3³D LCIF above ~ 2 Td

Simplified Scaling

$$\Delta N_{3^3D} \sim K_{ij}^e n_e \times \Delta N_{3^3P}$$

$$\frac{\Delta N_{3^3D}}{N_{3^3P}} \sim K_{ij}^e n_e$$

Observed Scaling of LCIF

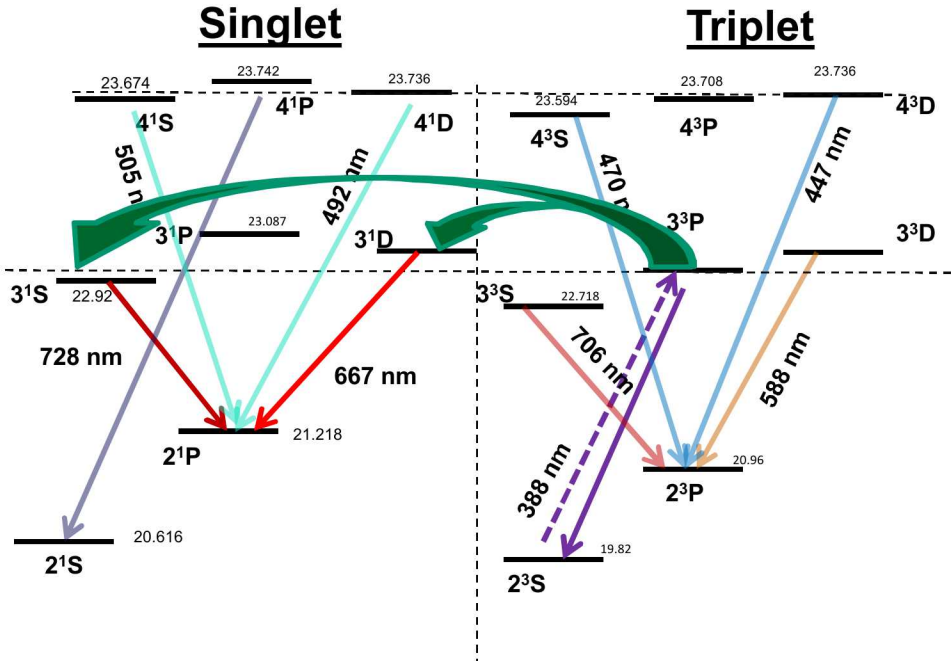


LCIF from 3³D scales linearly with electron density

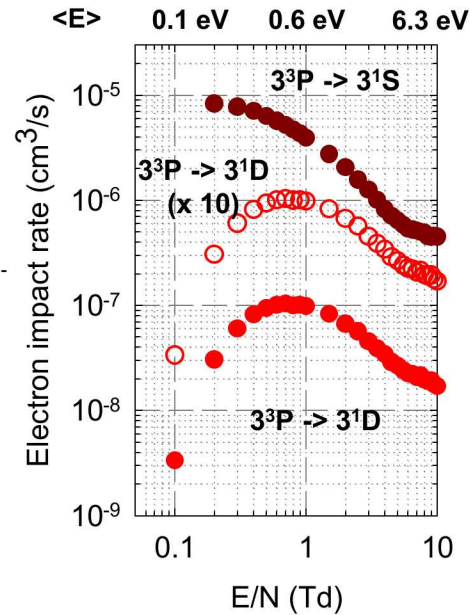
LCIF FROM SINGLET STATES BOUND E/N BELOW 1 Td

- Additional transitions near the laser excited 3^3P state are considered
 - LCIF emanating from singlet states are observed.

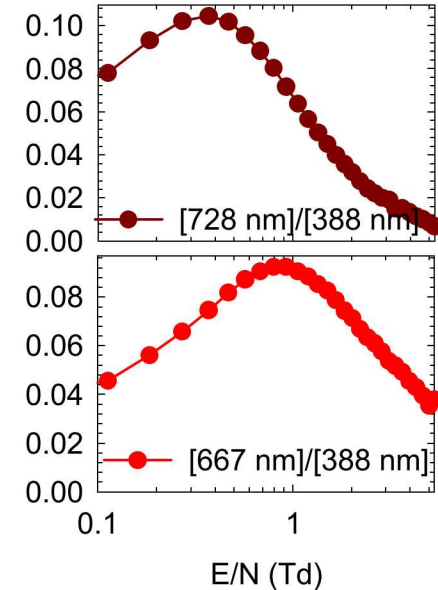
Pathway



Rates



Scaling of LCIF



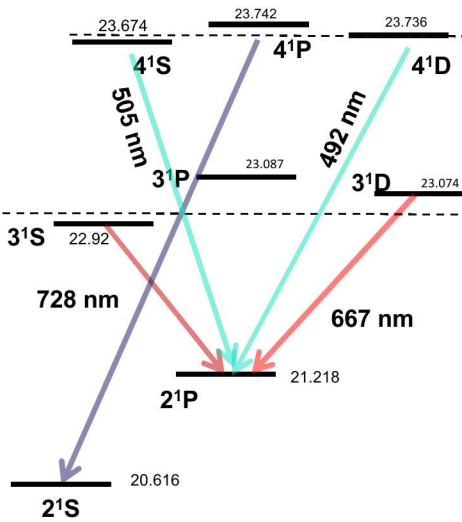
Emission from singlet states show similar scaling with E/N.

LCIF FROM 4³D BOUNDS E/N ABOVE 1 Td

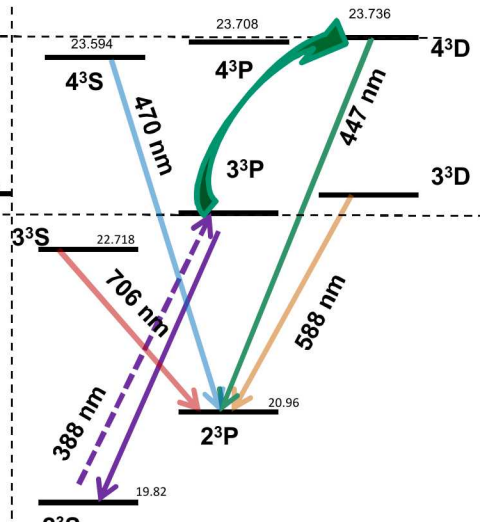
- LCIF emanating from states farther from laser excited 3³P state is observed.
 - LCIF from 4³D is discussed but other states are observed.

Pathway

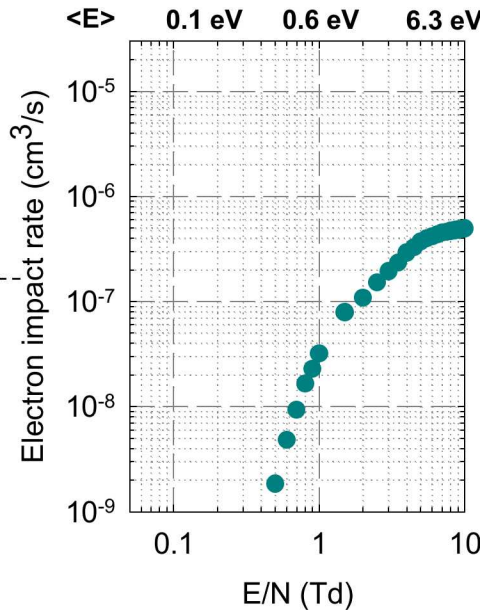
Singlet



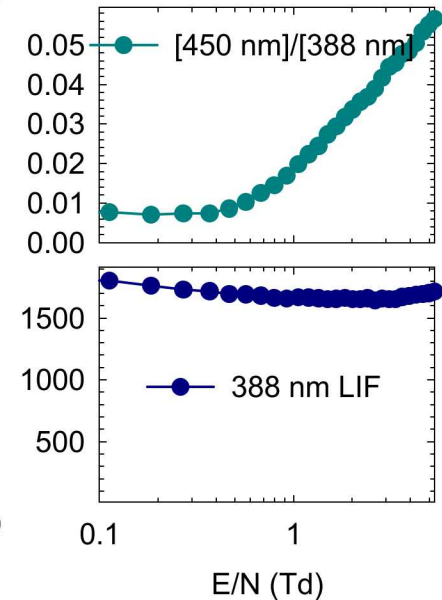
Triplet



Rates



Scaling of LCIF



Non-zero LCIF from 4³D at low E/N suggests that other factors contributing to signal (To be discussed later).

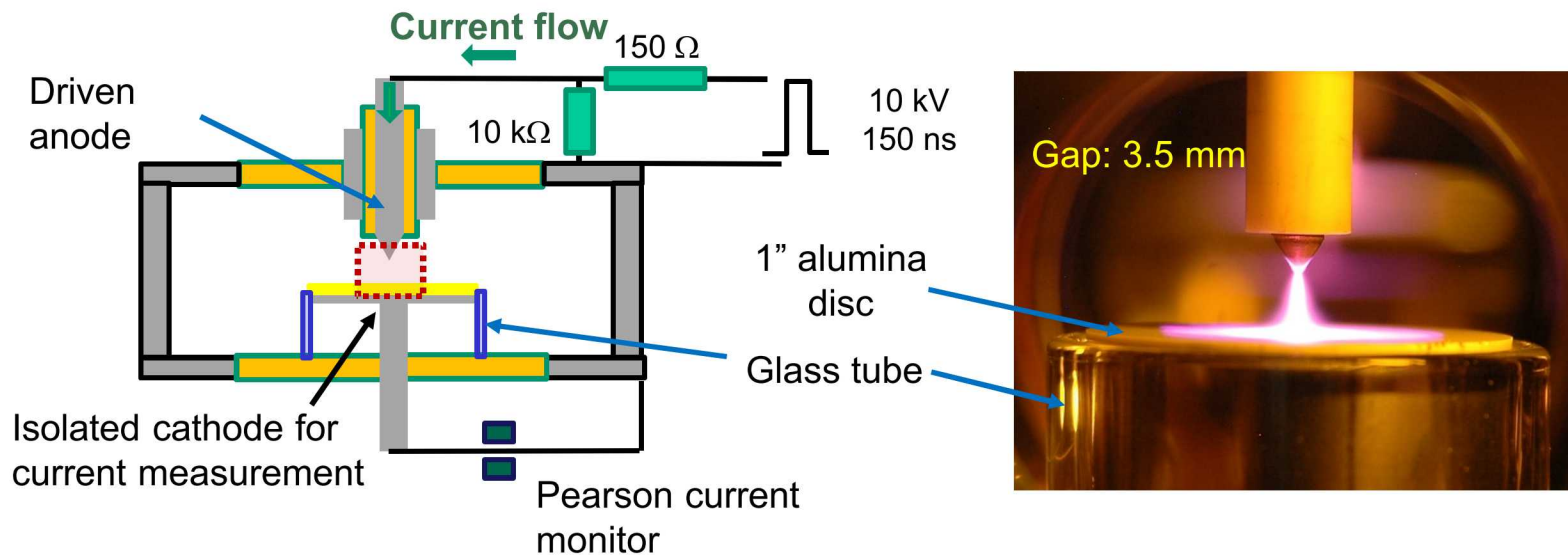
Part II: Application to various high pressure discharges

- Part I: Overview of laser-collision induced fluorescence (LCIF)
 - LCIF Concepts and challenges with LCIF at higher pressures
 - Extension of LCIF and demonstration of key LCIF trends
- **Part II: Application to various discharge systems of interest**
 - **Interrogate structured and dynamic systems**

Highlight interesting observations enabled with application of the LCIF to these systems

APPLICATION OF LCIF ON POINT-PLANE GEOMETRY TO INTERROGATE NEAR SURFACE

- Interested in the surface and near-surface plasma properties
 - Very challenging environment to access and interrogate.

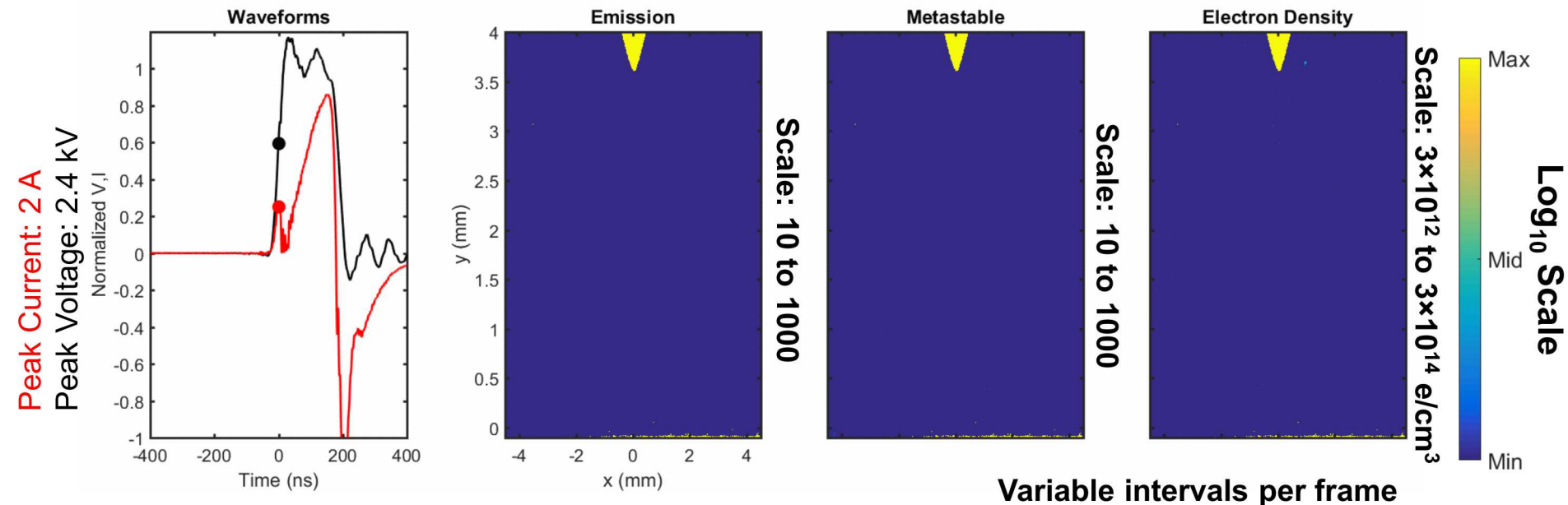


Dynamic environment to test and demonstrate LCIF method.

SPATIAL AND TEMPORAL EVOLUTION OF ATMOSPHERIC PRESSURE PLASMA IS OBSERVED

- LCIF method captures various phases of plasma formation.
 - Initiation (20 ns) -> Discharge (130 ns)-> Afterglow (~500 ns).

Entire Discharge Event

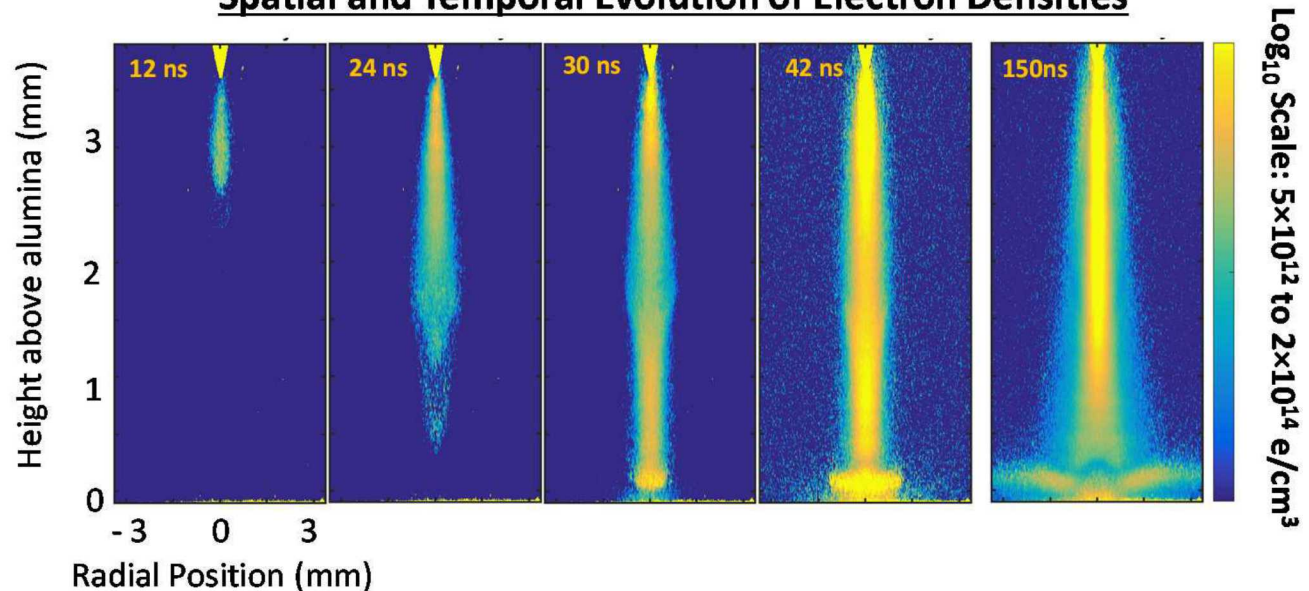
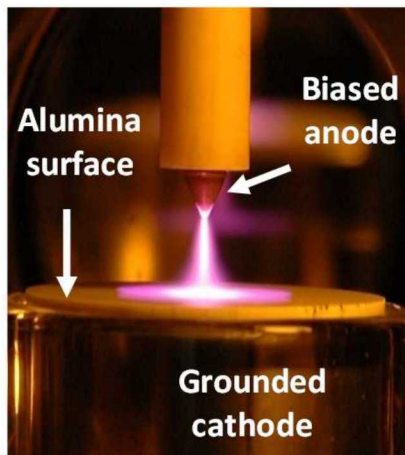


Several interesting aspects of the discharge will be discussed in context of trying to understand discharge and scrutinizing LCIF.

POINT TO PLANE DISCHARGE WAS FIRST SYSTEM THAT WAS STUDIED WITH LCIF

- LCIF provides high spatial and temporal resolution of the ionization wave formation process
 - Diagnostics provide access to a really challenging environment.

Spatial and Temporal Evolution of Electron Densities

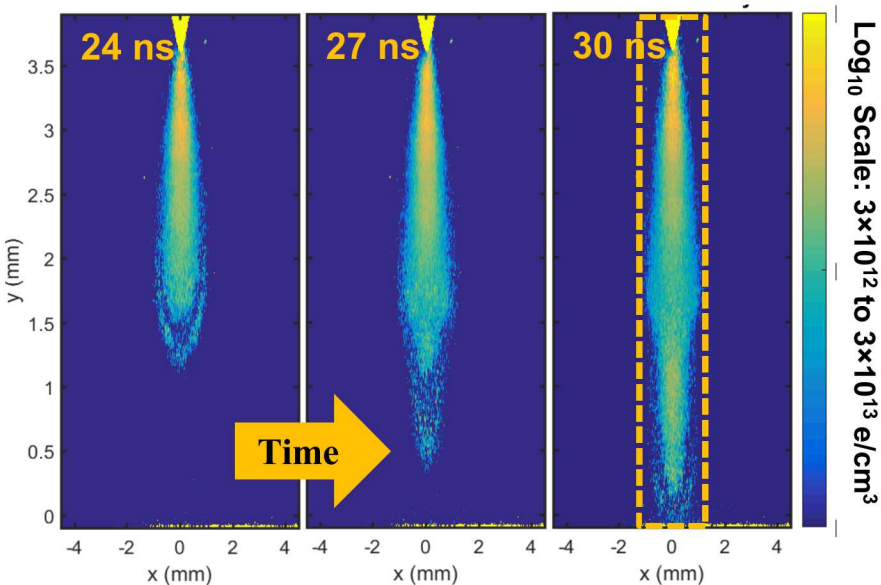


Test case used for proof of principle demonstration... Observations could seed further collaboration on plasma-surface interactions.

PROPIGATION OF FIW ACROSS GAP CHANGES MODE NEAR SURFACE

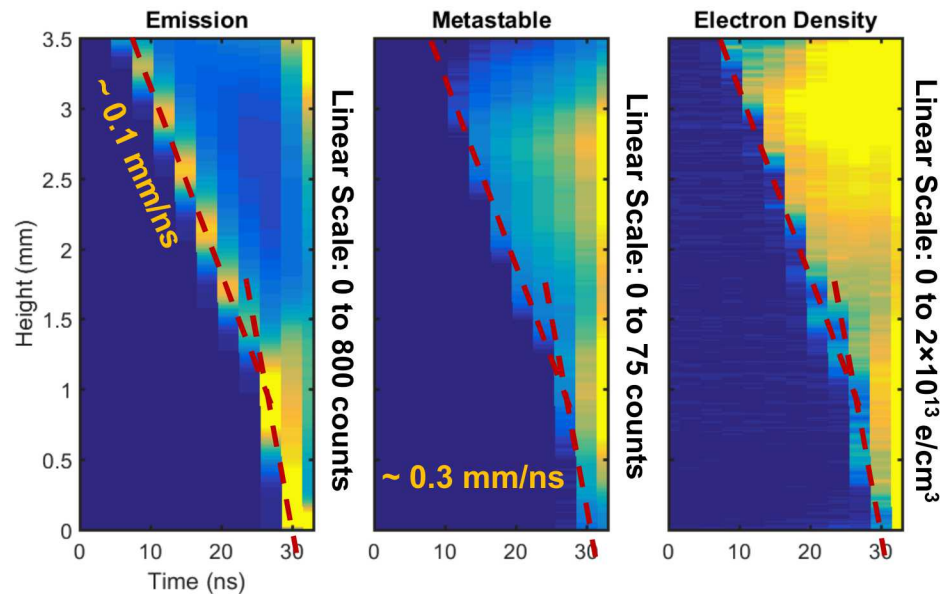
- Initiation ionization wave picks changes velocity near the surface.
 - Initial radially expanding “slow” (~ 0.1 mm/ns) front.
 - Directed “fast” (~ 0.3 mm/ns) front near surface.

Snapshots of Electron Density



ICCD gate of 3 ns

Axial Streak Profiles of Initiation



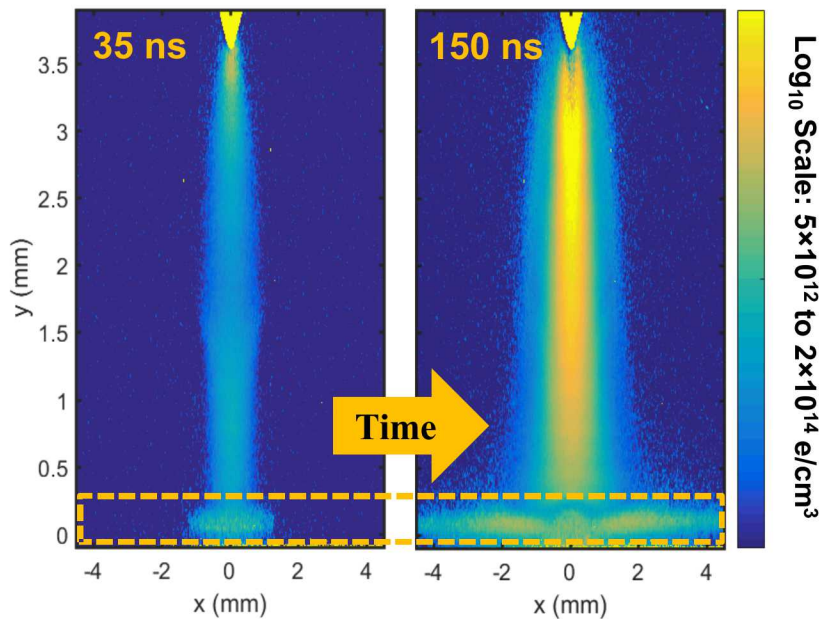
Averaged area: $0 \mu\text{m} \pm 50 \mu\text{m}$ on axis

Plasma propagation and formation times require “fast” diagnostics to capture.

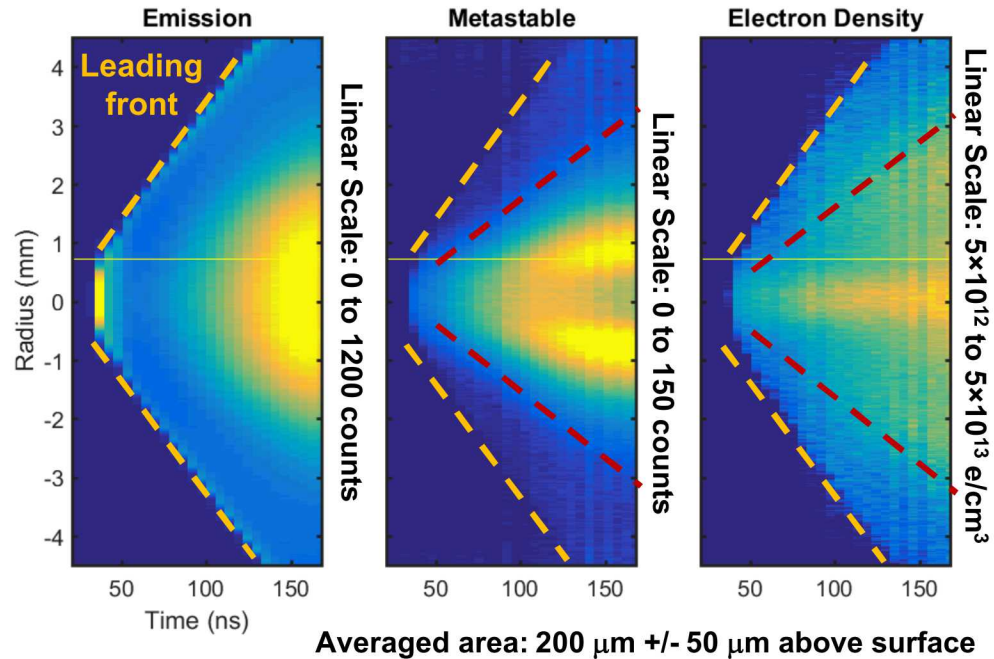
FORMATION OF A SURFACE IONIZATION WAVE IS OBSERVED WITH LCIF

- Multiple fronts are observed as the wave spreads.
 - Density $\sim 2 \times 10^{13}$ e/cm³ at leading front, slowly builds later.

Snapshots of Electron Density



Radial Streak Profiles of Surface Wave

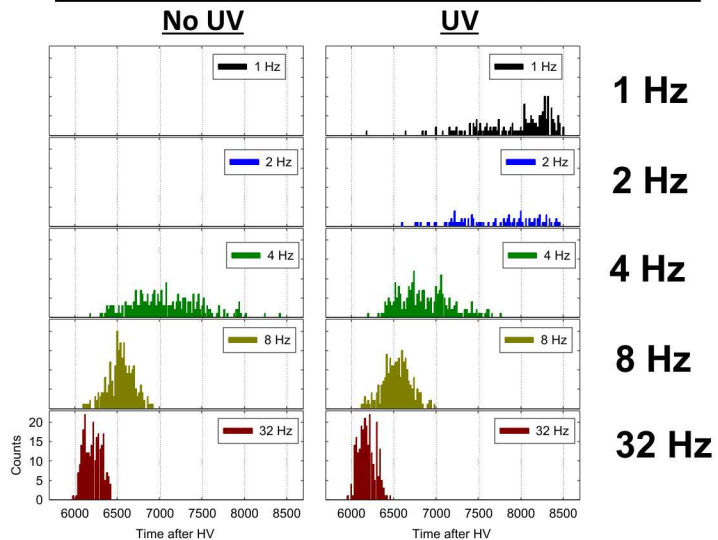


Double layer "like" partitioned structure emerges during active plasma discharge.

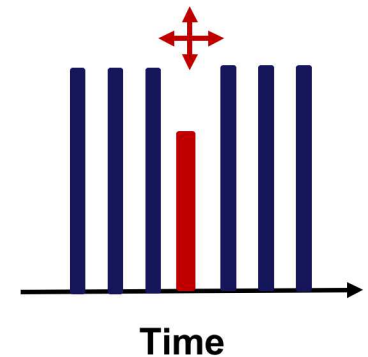
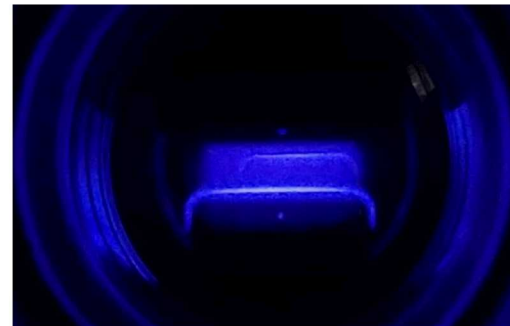
PLANE TO PLANE DISCHARGE UTILIZED TO STUDY “MEMORY EFFECTS” ON PLASMA

- Collaborative effort with P. Bruggeman and M. Simeni Simeni (U. Minnesota) to study role of “memory effects” on plasma discharge formation.
 - How is the plasma generating process influenced by residual species?
- Utilize plane-plane discharge to reduce plasma to quasi 1D to model observations and simplify measurements
 - Employ modified double pulse method to induce “memory effects”

Illustration of memory effect



Setup and Methodology

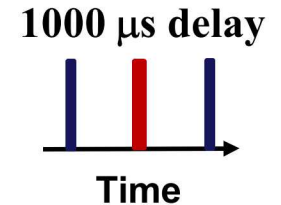


Nominal rate (minus one pulse)
Red pulse that fill in the gap

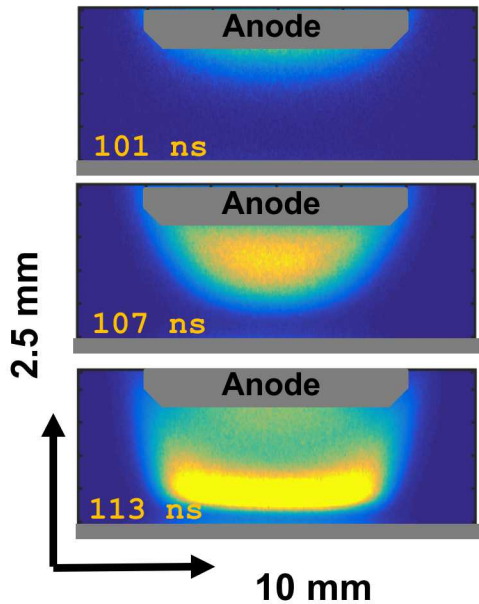
Develop methods to induce and interrogate this memory effect.

PLASMA GENERATION IS MONITORED AS A FUNCTION OF DELAY

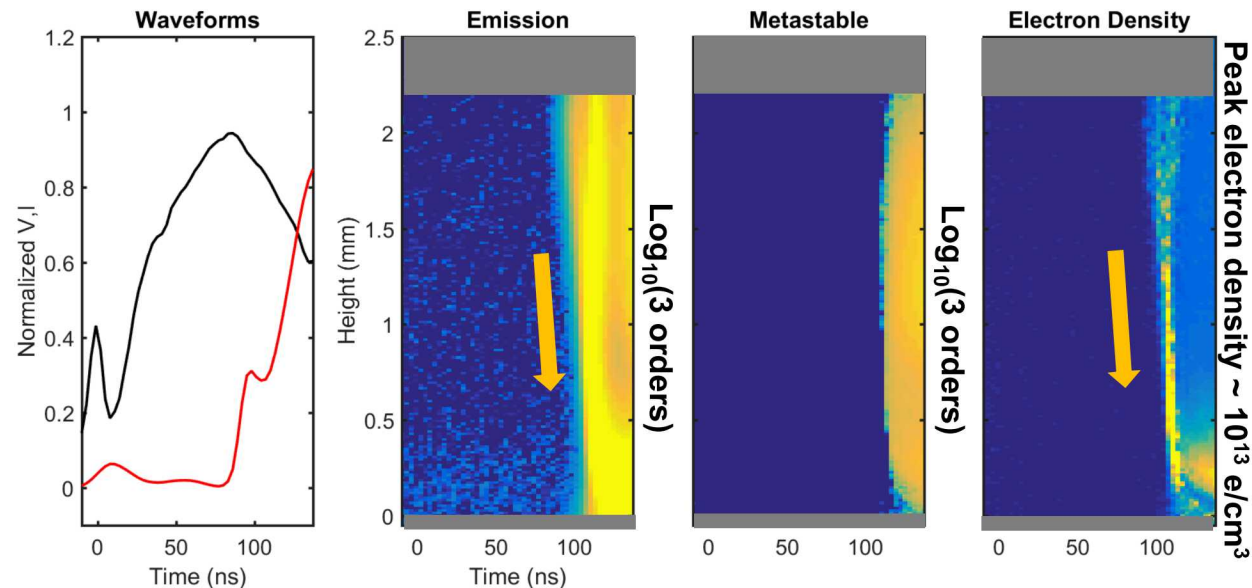
- Nominal 1 kHz rate studied to provide base line.
 - 1.3 kV, 150 ns pulse, 200 Torr Helium.
 - 1000 μs delay (1 kHz nominal rate).



2D Plasma Emission



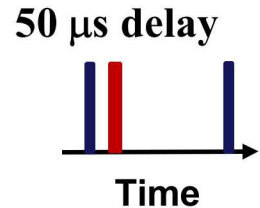
Averaged Streak Profile



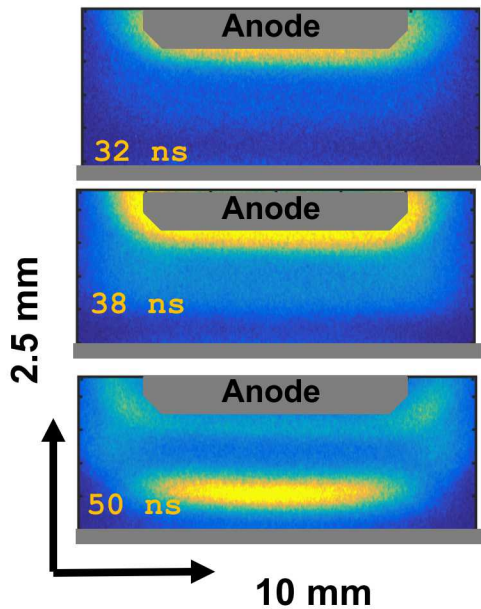
Cathode directed ionization wave is observed.

PLASMA GENERATION IS MONITORED AS A FUNCTION OF DELAY

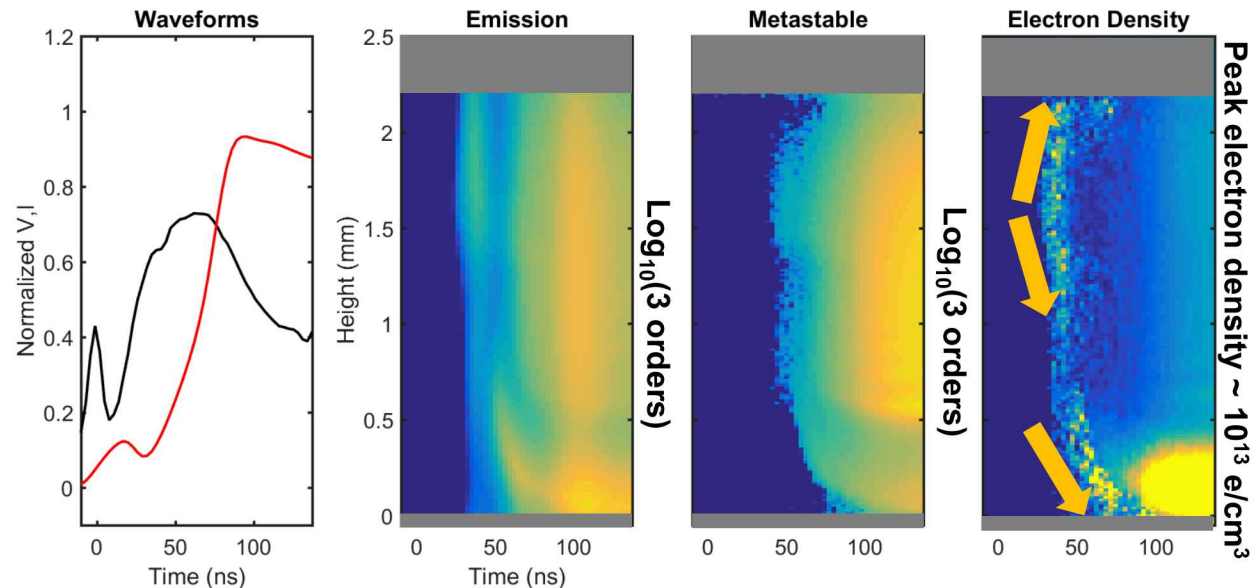
- Modified 1 kHz rate with one pulse offset in time
 - 1.3 kV, 150 ns pulse, 200 Torr Helium
 - 50 μs delay.



2D Plasma Emission



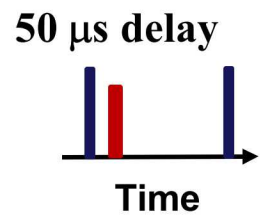
Averaged Streak Profile



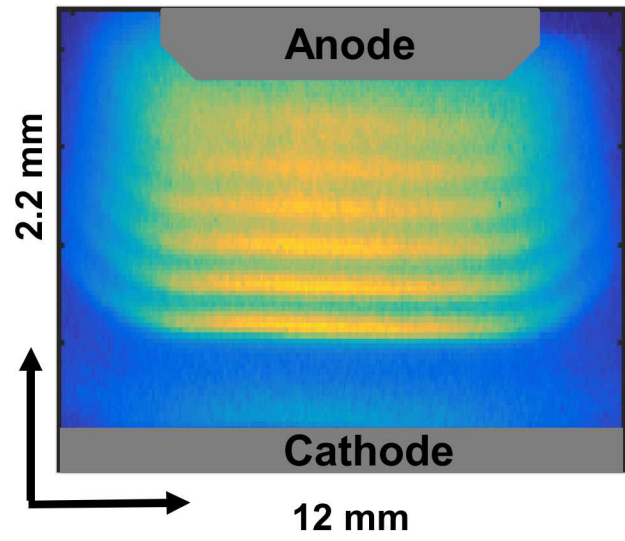
Plasma generation is more volumetric and plasma shows more structure

PLASMA DISPLAYS STRIATED STRUCTURE

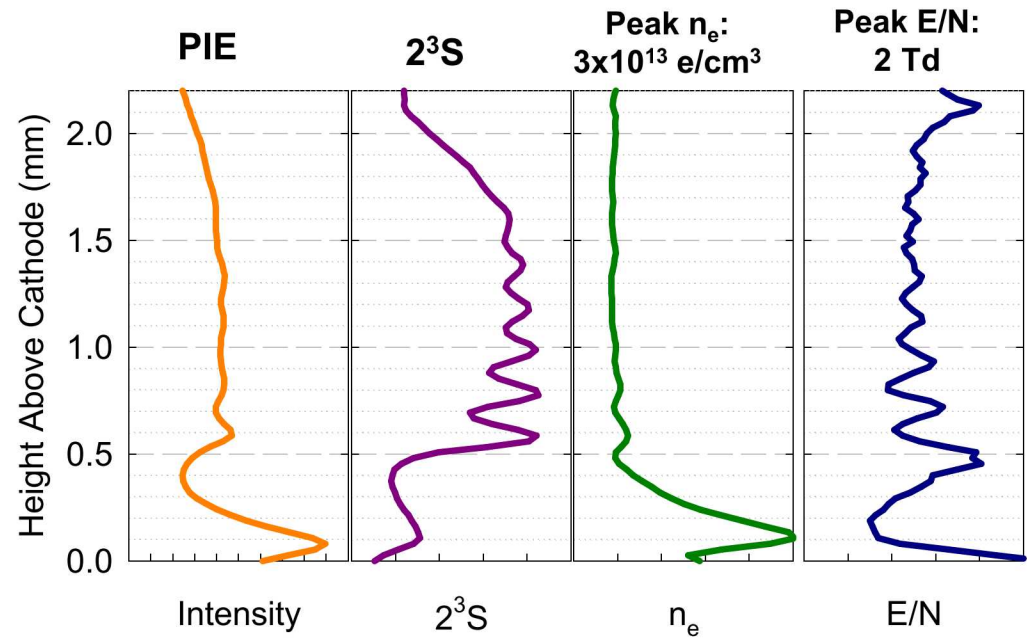
- Under certain drive conditions striated plasma is observed
 - 1.0 kV, 150 ns pulse, 200 Torr Helium
 - 50 μ s delay.



Metastable Distribution



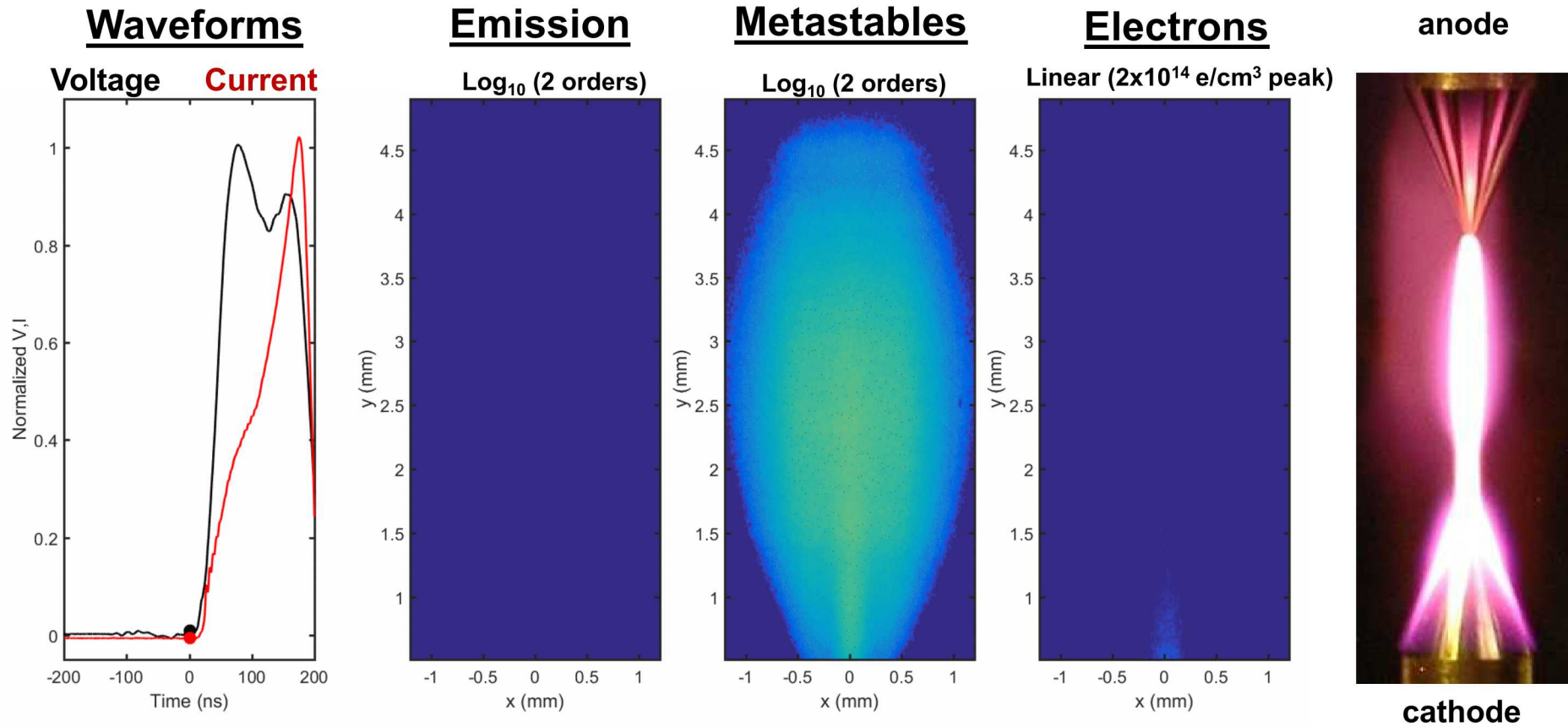
On-center Profiles



Structures in metastable distribution is not easily observable by emission...

DYNAMICS OF PIN TO PIN DISCHARGE OBSERVED

- Measured evolution of plasma formation during second pulse.
 - Initial peak electron density of 2×10^{13} e/cm³ (2 μ s afterglow)

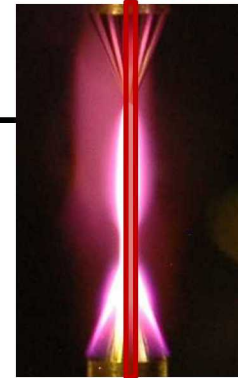


LCIF provides two-dimensional plasma structure as the plasma transitions from a glow to an arc

UTILIZATION OF LCIF TO UNDERSTAND DISCHARGE EVOLUTION

anode

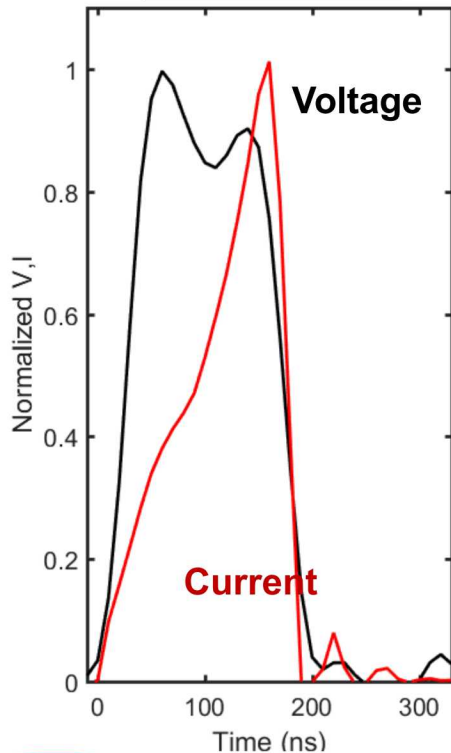
Averaged domain



cathode

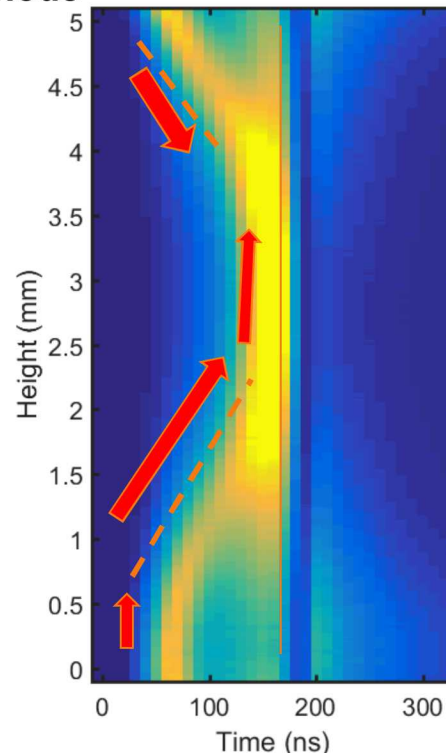
- Streak-like images of plasma initiation to examine dynamics of plasma formation.
 - Axial profiles at center of discharge.

Waveforms

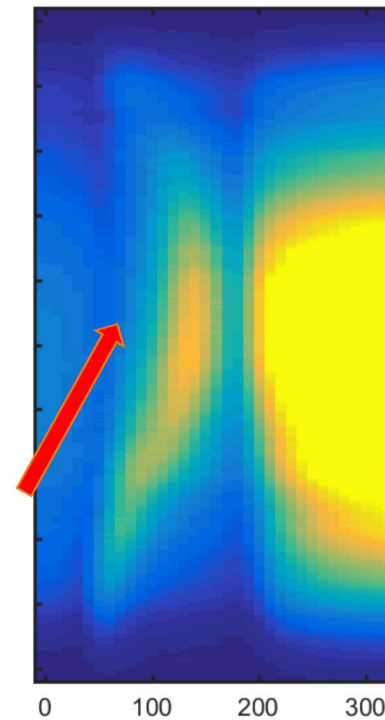


Powered anode

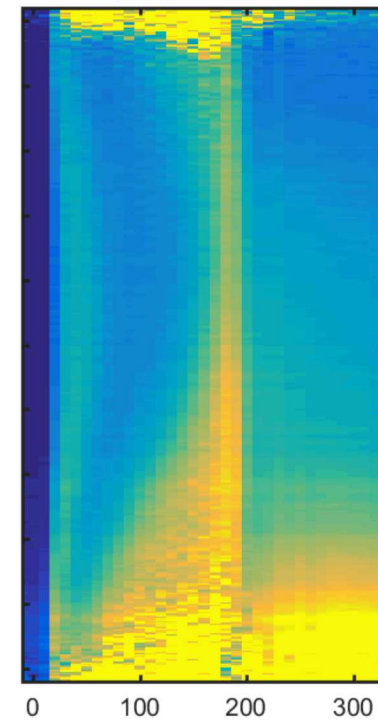
Emission



Metastables



Electrons



Grounded cathode

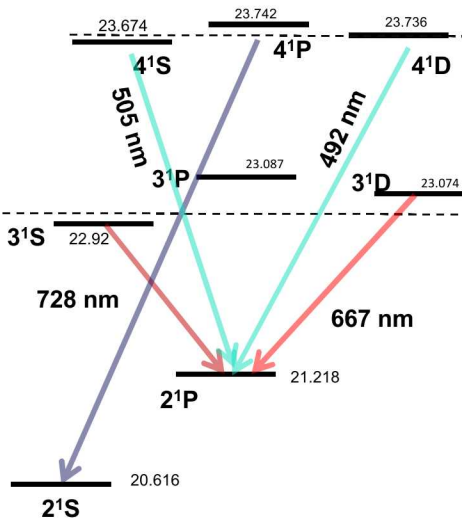
LCIF captures spatial and temporal evolution of plasma formation.

LCIF FROM 4³D BOUNDS E/N ABOVE 1 Td

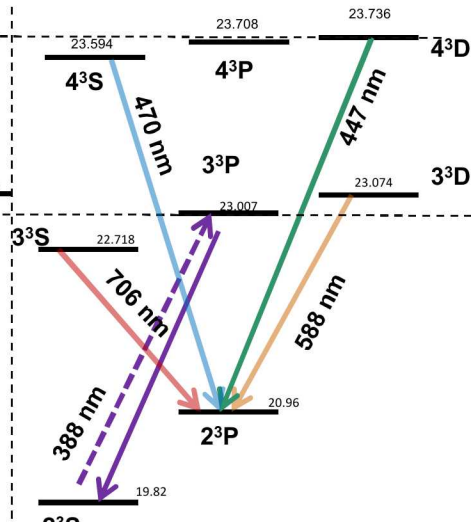
- LCIF emanating from states farther from laser excited 3³P state is observed.
 - LCIF from 4³D is discussed but other states are observed.

Pathway

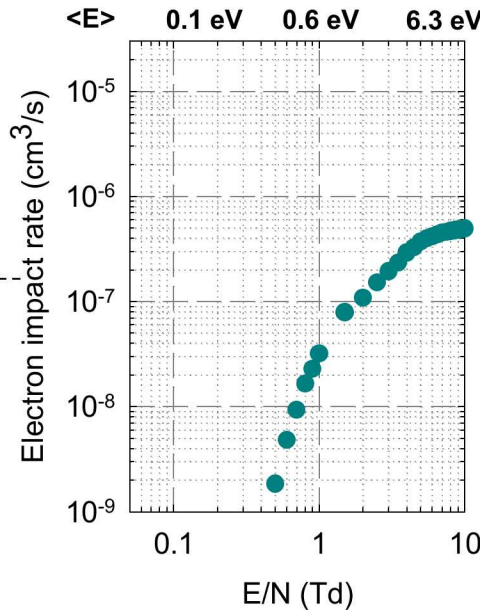
Singlet



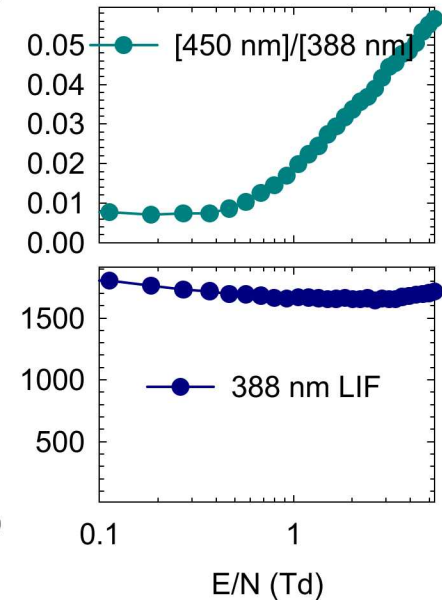
Triplet



Rates



Scaling of LCIF

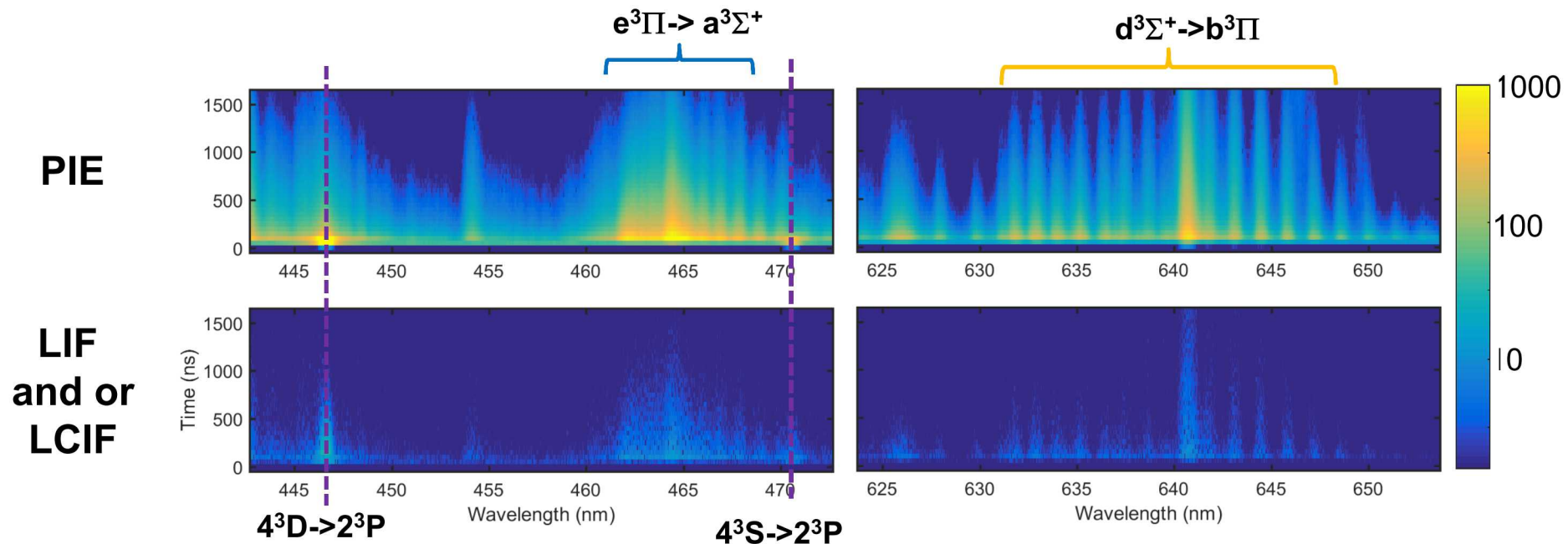


Non-zero LCIF from 4³D at low E/N suggests that other factors contributing to signal.

SPECTRAL SURVEY INDICATES THAT MOLECULAR HELIUM IS BEING GENERATED

- Plasma induced emission (PIE) measures species present in the plasma.
 - LIF and/or LCIF is also observed emanating from molecular species.

Temporal Evolution of Selected Spectrum

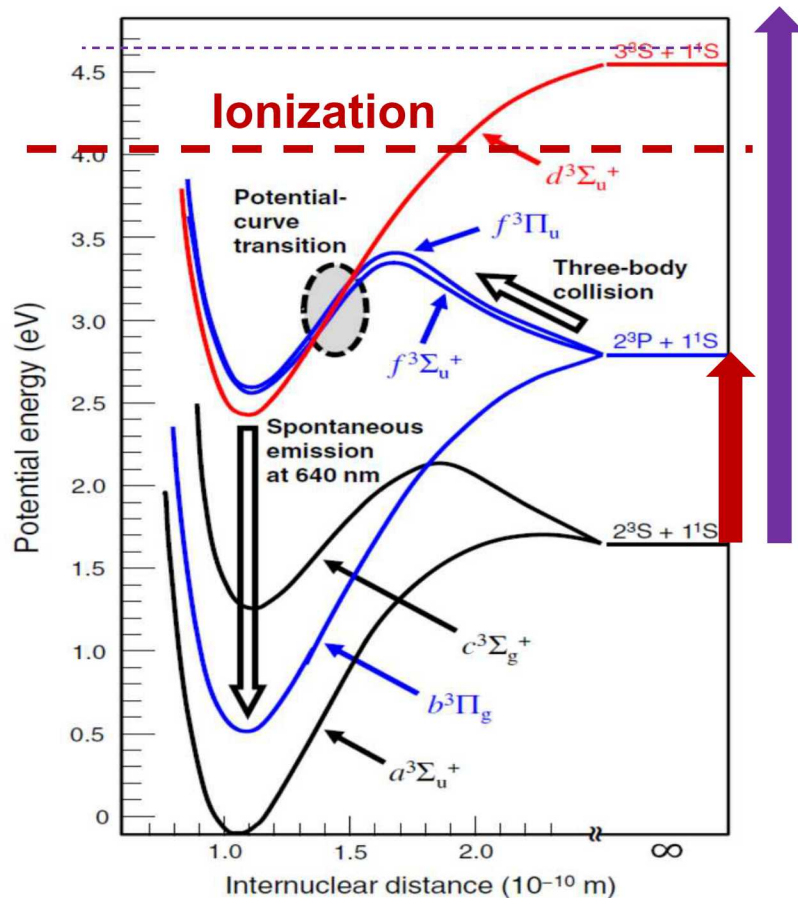


Several other molecular bands from both triplet and singlet states are (potentially) identified.

MOLECULAR FORMATION HAS BEEN OBSERVED IN EARLIER STUDIES

- Earlier studies identified increased molecular emission after laser excitation
- Urabe excited 2^3P with CW laser
 - Observed molecular emission around 641 nm
- Frost excited 3^3P with nanosecond laser
 - Observed several molecular transitions.
 - Fluorescence from molecular lines had similar temporal evolution of laser excited 3^3P state.
- Recent studies by Carbone et al discussed role of molecules in high pressure plasma
 - Stated open questions about how molecules form
 - Association vs. Ionization and recombination.

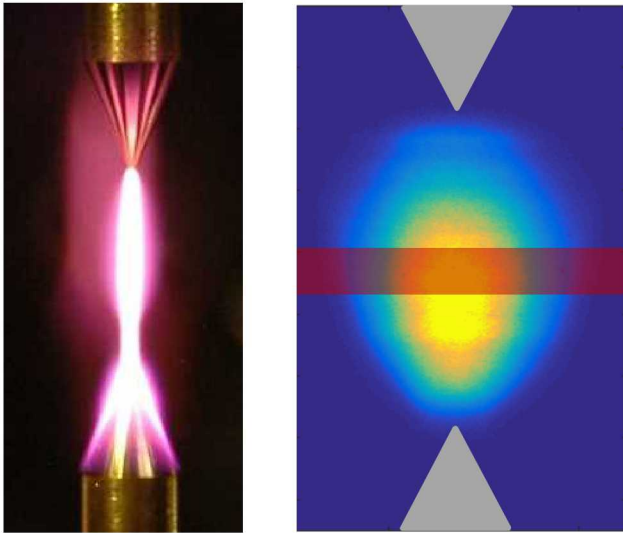
Uncertainty exist about specific pathways for molecular emission.



MOLECULAR FORMATION SHOWS STRONG DEPENDENCE ON DISCHARGE CONDITIONS

Show other snapshots demonstrating molecule formation

Plasma configuration



E/N Dependence

E/N: 0.1 -> 4 Td

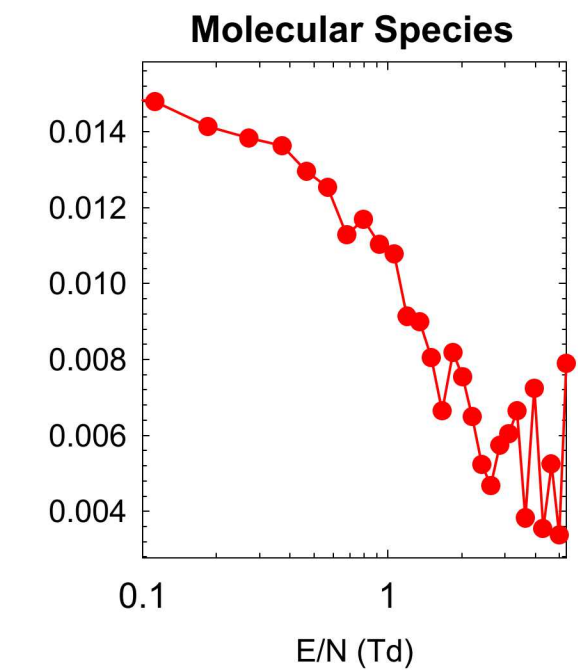
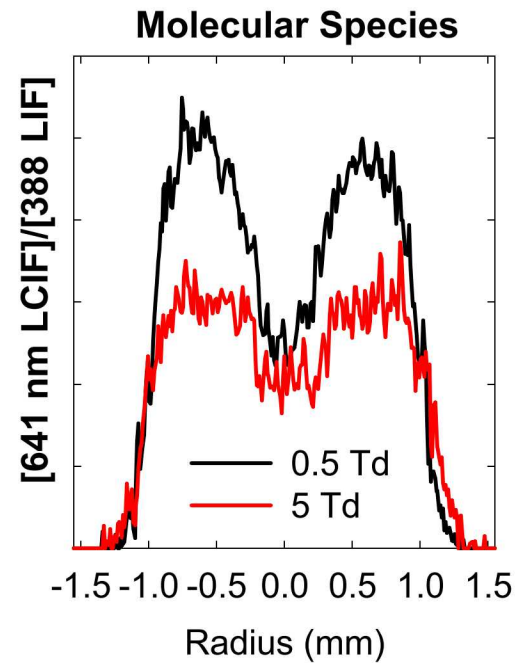
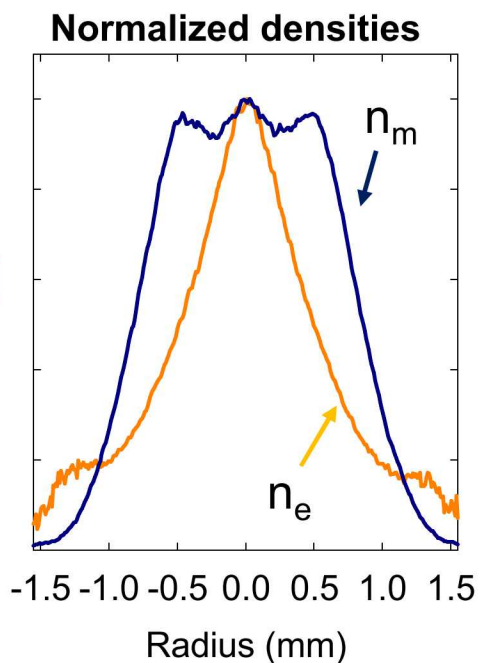
Recast this using selected profiles – Fixed Density, Fixed Metastable

MOLECULAR FORMATION SHOWS STRONG DEPENDENCE ON DISCHARGE CONDITIONS

- Images of LCIF from molecular transition $d^3\Sigma^+ \rightarrow b^3\Pi$ were obtained and analyzed as functions of E/N in afterglow.
 - Metastable species and electron densities are expected to remain fixed

Radial Distribution

E/N Dependence



Do these observations suggest change in associative pathways?

CONCLUSIONS

- **Ultra-fast LCIF shows promise for interrogating high pressure plasma systems.**
 - **Outlined pitfalls that might be encountered at higher-pressure systems.**
 - **Can be extended to other systems of interest (Ar, N,...).**
- **Preliminary LCIF results were published earlier this year**
 - **E.V. Barnat and A. Fierro, J.Phys.D:Appl. Phys. 50 (2017) 14LT01**

Thank you for your attention!

*This work was supported by the Department of Energy Office of Fusion
Energy Science
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AUXILARY SLIDES

NEUTRAL INTERACTIONS KEY SOURCE OF INCREASED COMPLEXITY

- Neutral-impact redistribution can play dominant role at higher pressures
 - More-types and evolving nature of neutrals (dimers).
 - “Book keeping” can require sophisticated models.
 - Uncertainties in species and cross-sections limit accuracy.

Neutral mixing

$$\frac{dN_j}{dt} = \sum_k \left[\sum_{i \neq j} K_{ikj}^a N_i - \sum_{i \neq j} K_{jki}^a N_j \right] N_k$$

Bounds on detection

$$\frac{\Delta N_{Electrons}}{\Delta N_{Neutrals}} \sim \frac{K^e n_e}{K^N n_0} \sim 1$$
$$n_e \sim \frac{K^N}{K^e} n_0 \sim \frac{10^{-11}}{10^{-5}} n_0$$

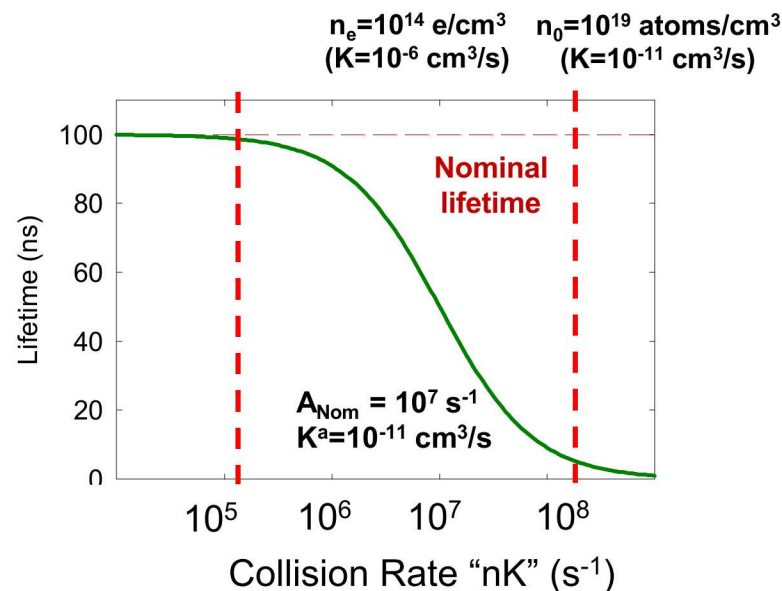
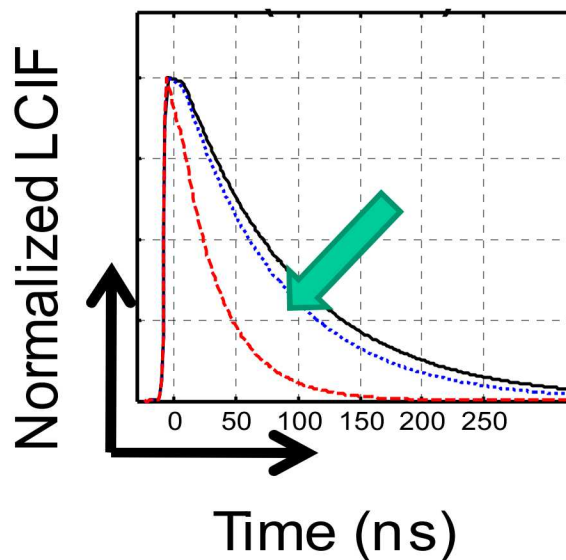
Limit $\sim 10^{13}$ e/cm³ at 1000 Torr

Neutrals are anticipated place limits on lower bound of electron detection.

LIFETIMES OF EXCITED STATES BECOME VERY SHORT AT HIGHER DENSITIES

- Physics of electron-impact redistribution is not expected to change at higher pressures.
 - Sheer number of electrons increase probability of redistribution.
 - Effective Lifetimes become reduced because of redistribution.

$$\frac{dN_j}{dt} \sim \left[\sum_{i \neq j} K_{ij}^e N_i \right] n_e$$

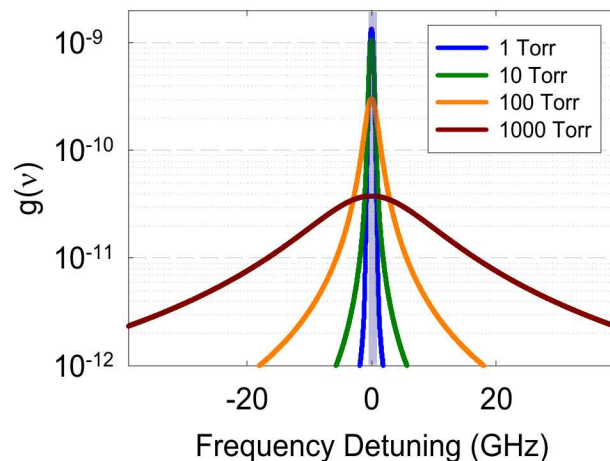


Lifetime of excited states are quite short (~5 ns) at target conditions.

ULTRASHORT-PULSE LASER IS USED FOR INITIATION LCIF EVENT

- Ti:Sapphire, regenerative laser used to generate excitation pulse.
 - Tuned amplifier to 780 nm – doubled in BBO for ~ 390 nm.
 - ~ 100 fs pulse with 10 nm bandwidth (~ 100 cm^{-1}).
- Short-pulse laser well suited to interrogate short lifetimes (< 10 ns) and broad absorption profiles (~ 1 nm) associated with high pressure.
 - Still realize “step-like” populating process.
 - Sample most or all of the probed states.

Anticipated absorption profiles



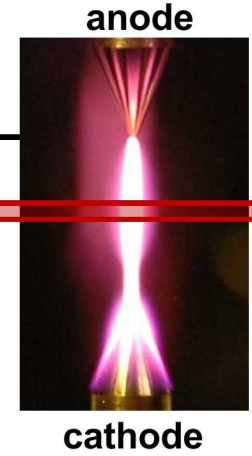
Estimates of linewidths

ns laser ~ 0.5 GHz or 0.1 cm^{-1}

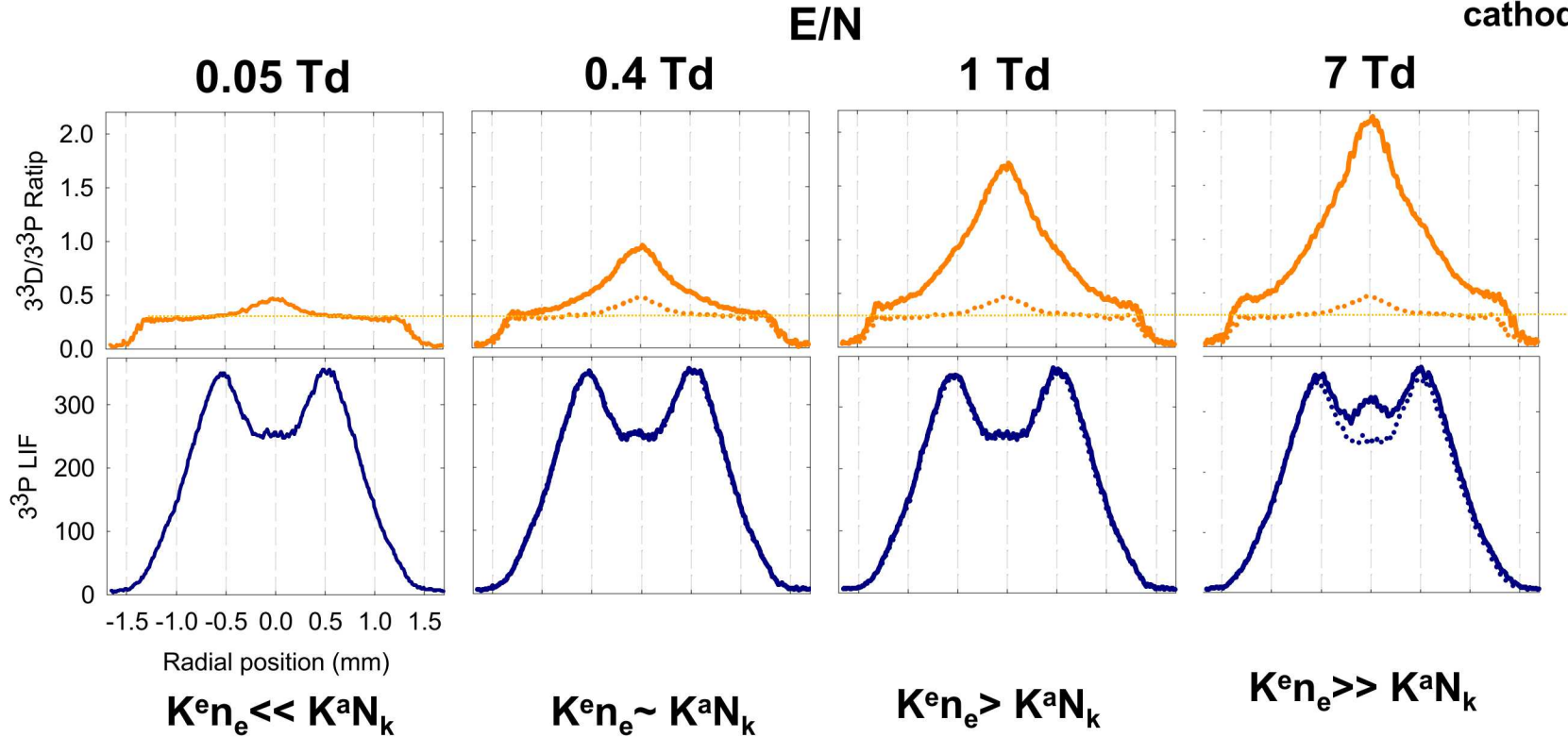
fs laser ~ 500 GHz or 100 cm^{-1}

Pressure Broadening ~ 0.01 GHz/Torr (He)

DEMONSTRATION OF SPATIAL RESOLUTION PROVIDED BY LCIF



- Radial structure observed for various E/N.
 - Initial peak electron density of 2×10^{13} e/cm³.
 - Measured $\sim 2 \mu\text{s}$ in afterglow of first voltage pulse.
 - Measured in first 20 ns of applied heating pulse.

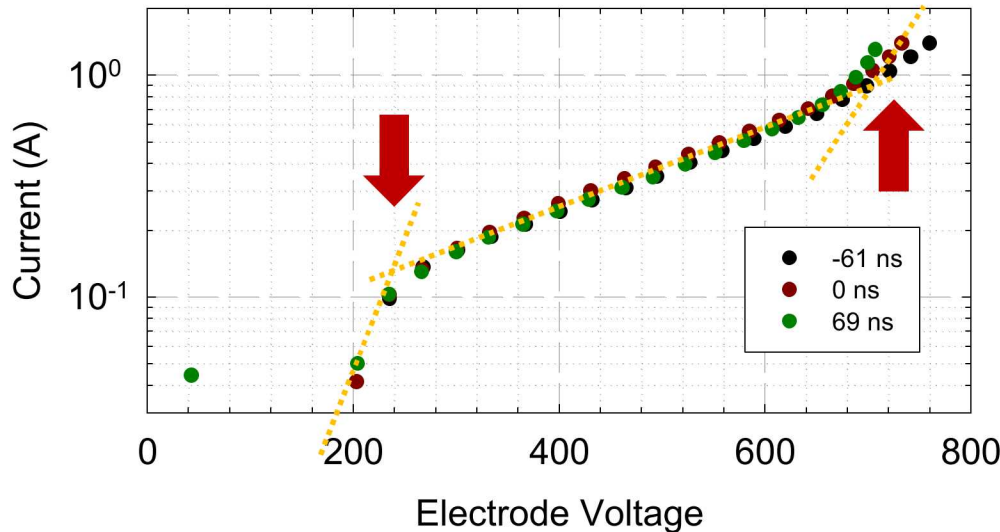


Observations consistent with initial assertions made

UTILIZE CURRENT-VOLTAGE TRENDS TO BOUND PLASMA PARAMETERS

- Published drift data (Phelps) is used to bound E/N with heating voltage.
 - Knees in current correspond to knees in drift velocities.
 - Electron density remains roughly constant at lower E/N values.

Extracted Current



Anticipated plasma parameters

$$n_e = \frac{m_e v_m N I}{e^2 N E A}$$

V	I	J	E/N	Vd	ne
V	A	A/cm ²	Td	cm/s	e/cm ³
225	0.1	3.18	0.1	1.50E+05	1.32E+14
700	0.6	19.10	5	1.00E+06	1.19E+14

Delay between first pulse and second pulse - Density

Magnitude of second voltage pulse – E/N