

ULTRAFAST LASER DIAGNOSTICS TO INTERROGATE HIGH PRESSURE PLASMA ENVIRONMENTS

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INTRODUCTION AND MOTIVATION: CHALLENGES AND OPPORTUNITIES

- Diagnostic development and implementation has been contribution SNL has made to PSC.
 - Used these methods to interrogate systems of interest.
- High pressure (~ 1 ATM and beyond) is challenging.
 - Higher densities.
 - Smaller length scales.
 - Shorter lifetimes.
 - Optically thicker environments.
- Ultrafast (UF) lasers show great potential for addressing some of these challenges.
 - Shorter timescales (<100 fs) to interrogate fast phenomenon.
 - Large bandwidth to pressure-broadened states.
 - High intensity to drive non-linear processes.

Overall goal is to match these challenges to potential opportunities offered by short-pulse lasers.

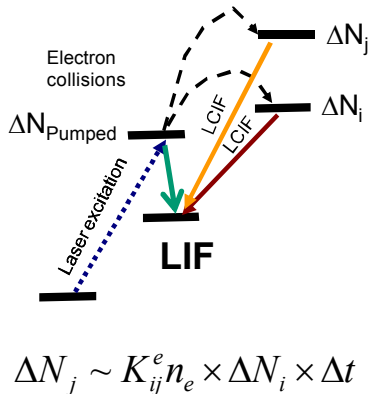
OVERVIEW OF RECENT RESEARCH EFFORTS: GOALS AND TARGETS

- **Short term goal: Applying LCIF to higher pressure plasmas.**
 - **LCIF: Laser-collision induced fluorescence.**
- **Overview of LCIF and challenges at higher pressures.**
 - **Anticipated challenges understanding observed signals.**
- **Generating and controlling higher pressure discharges.**
 - **Double pulse method for generating and manipulating plasma.**
- **Implementing short-pulse for generating LCIF.**
 - **100 fs broad-band pulse for excitation.**
- **Proof-of-principle demonstration of LCIF.**
 - **Identify LCIF and calibrate n_e , E/N dependence.**
 - **Demonstrate spatial-temporal evolution of 400 Torr He filament.**
- **Longer term goal: Investigate short-pulse methods beyond LCIF.**
 - **Gas-phase and surface sensitive methods.**

SHORT(ER) TERM GOAL IS TO EXTEND LCIF METHOD TO HIGH(ER) PRESSURE

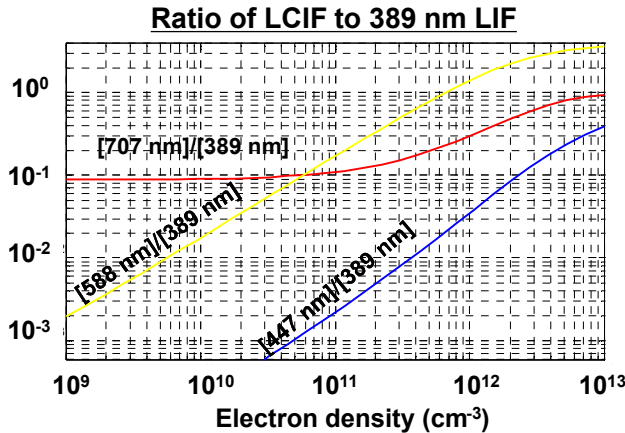
- **LCIF: Laser-collision induced fluorescence used to assess n_e , T_e .**
 - Laser excitation to known state.
 - Collisional redistribution to near-by states.

LCIF Concept

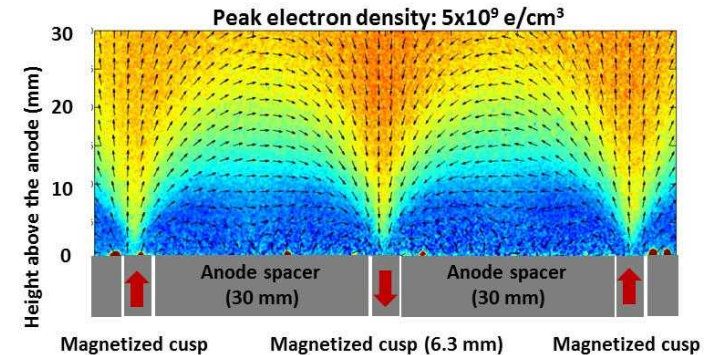


$$\text{Photons emitted} \sim A_{jk} \times \Delta N_j \times \Delta t$$

Observed Scaling



Implemented method



Once an atomic system is understood LCIF is powerful method.

OVERVIEW OF CHALLENGES OF EXTENDING LCIF TO HIGH PRESSURE

- Observed LCIF is superposition of several complex processes.
 - In general need a good model to describe the interaction of these processes on redistribution.
- Physics at lower pressures simplify much of the complexity.
 - Simplifications likely not to be so forthcoming at higher pressures.

"Electron mixing"

"Neutral mixing"

"Photon mixing"

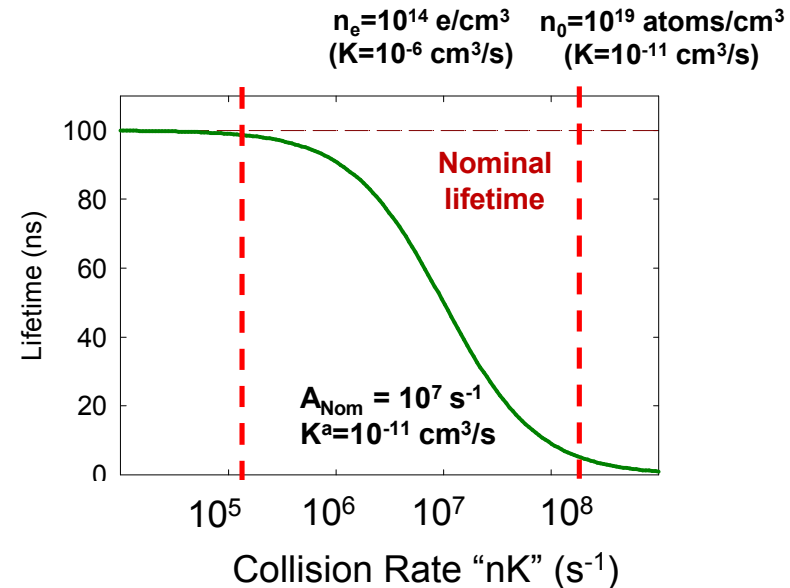
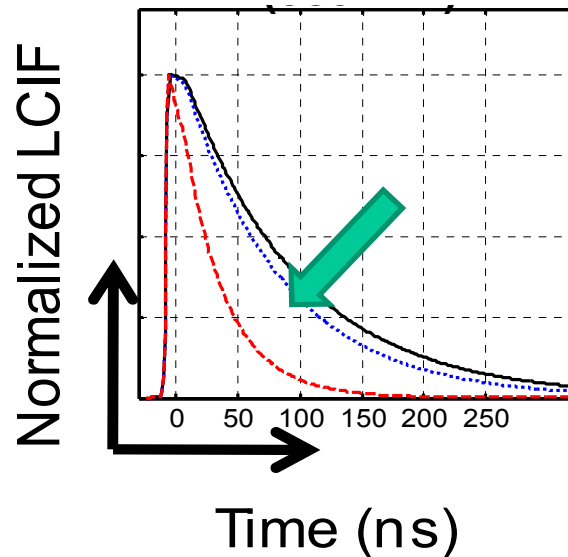
$$\frac{dN_j}{dt} = \left[\sum_{i \neq j} K_{ij}^e N_i - \sum_{i \neq j} K_{ji}^e N_j \right] n_e + \sum_k \left[\sum_{i \neq j} K_{ikj}^a N_i - \sum_{i \neq j} K_{jki}^a N_j \right] N_k + \left[\sum_{i > j} A_{ij} N_i - \sum_{i < j} A_{ji}^j N_j \right]$$

Considerable risk in porting this to higher pressures.

LIFETIMES OF EXCITED STATES BECOME VERY SHORT AT HIGHER ELECTRON 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.

NEUTRAL INTERACTIONS FURTHER COMPLEXITY

- Neutral-impact redistribution can play dominant role at higher pressures
 - Neutral collisions limit lower bound on electron detection.
 - More-types and evolving nature of neutrals (dimers).
 - “Book keeping” can require sophisticated models.

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

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 to play an increasingly important role

PHOTON TRANSPORT FURTHER COMPLICATES INTERPRETATION OF LCIF

- Tracking photon transport becomes much more important.
 - Short lifetimes become < 10 ns.
 - Higher densities lead to optically thick environments.
 - Radiation transport needs sophisticated models.

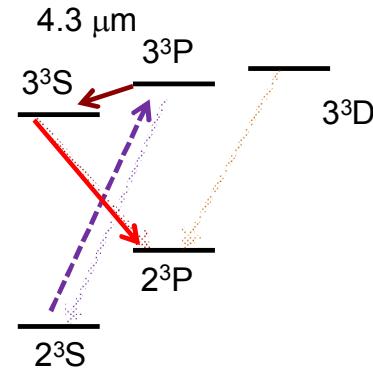
$$\frac{dN_j}{dt} = \left[\sum_{i>j} A_{ij} N_i - \sum_{i<j} A_{ji}^j N_j \right]$$

Scaling of MFP

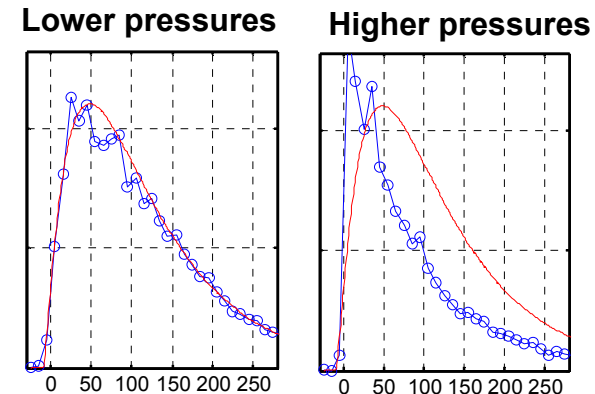
$$\tau = L / l_{mfp} \approx f_{nm} \lambda (Mc^2 / kT_A)^{1/2} N_A L \ll 1$$

Transition	f_{nm}	τ
$3^3P \rightarrow 2^3S$	0.064	> 1
$4^3P \rightarrow 2^3S$	0.02	~ 1

(Assuming $L=1$ mm and $N_A \sim 10^{14}$ absorbers/cm³)



Radiation transport



May need to account for photon trajectories and local environment.

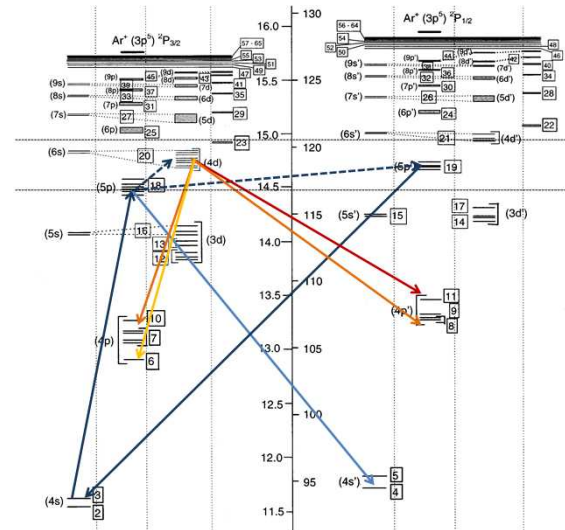
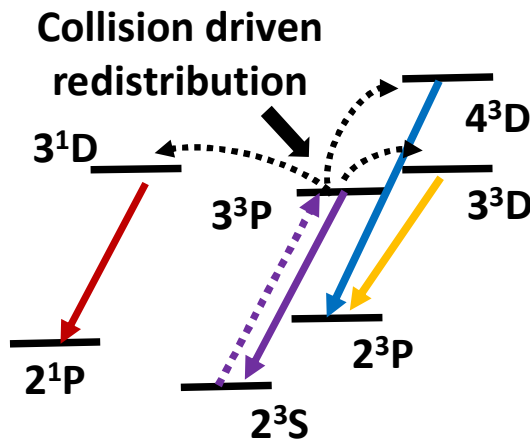
STRATEGIES USED TO DEVELOP LCIF AT HIGHER PRESSURES

- Helium is utilized as interrogated species.
 - Relatively “simple” system to start with.
 - Relevant workhorse at higher pressures !
- Short-pulse (<100 fs, ~ 10 nm) laser excitation.
 - Excitation times \ll decay times to simplify interpretation.
 - Curious to see if it would even work!
- Generation and manipulation of plasma in controlled environment.
 - Stable at target pressures (~ 1 ATM).
 - Well characterized and “easily” manipulated.

Much of the work was built on previous experience gained at lower pressures.

HELIUM IS RATHER “SIMPLE” SYSTEM THAT HAS BEEN WELL STUDIED

- Earliest LCIF development and implementation was in helium.
 - Isolated states and “well known” cross-sections between states.
- Later developments were made in argon.
 - Much more spectroscopically dense and unknown.

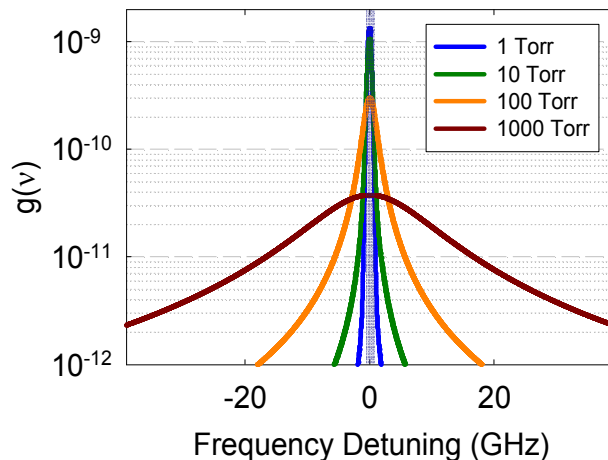


Start simple as higher pressure plasmas likely to add own layers of complexity.

SHORT-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

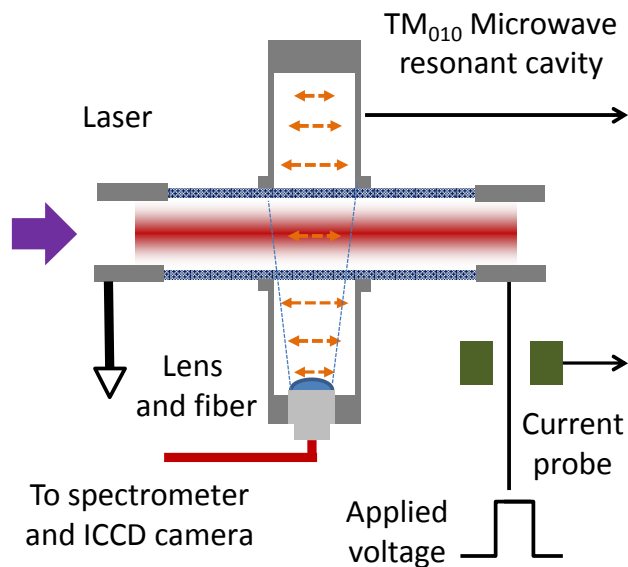
Pressure Broadening ~ 0.01 GHz/Torr (He)

Short pulse enables access to all of the pressure-broadened line

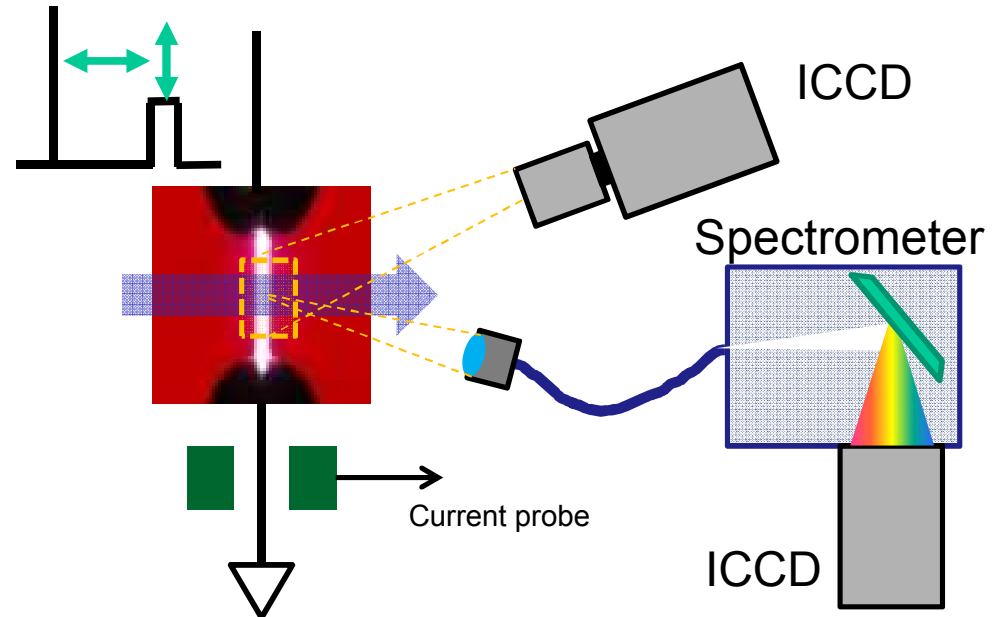
KEY CHALLENGE: GENERATING AND MANIPULATING WELL CHARACTERIZED PLASMA

- Previous experience in developing LCIF indicated that setup is key.
 - Double pulse method to separate generation and interrogation.
 - Microwave resonant cavity to assess densities.

Low pressure configuration



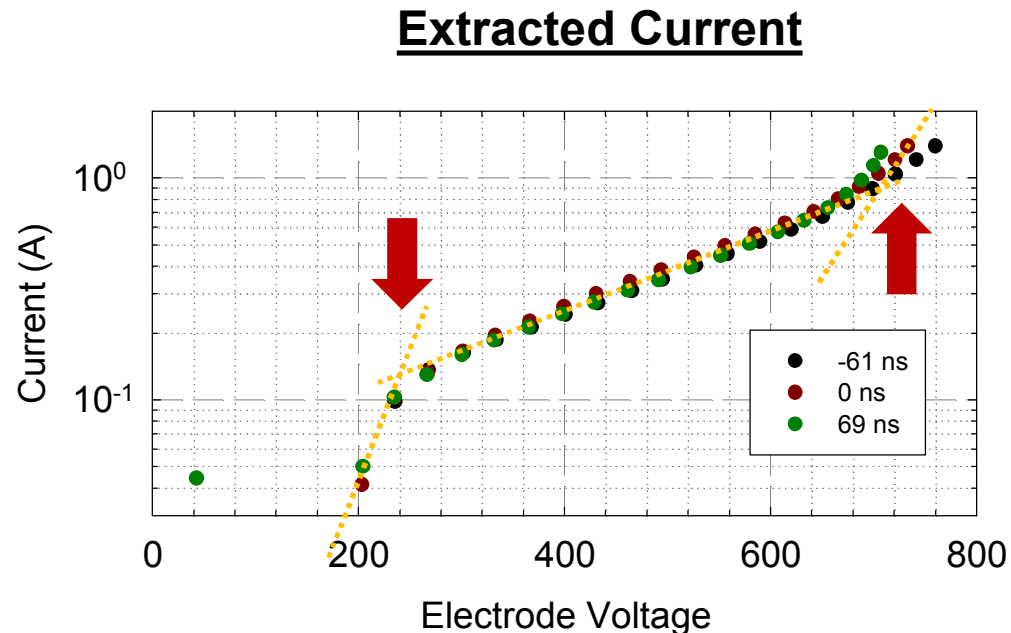
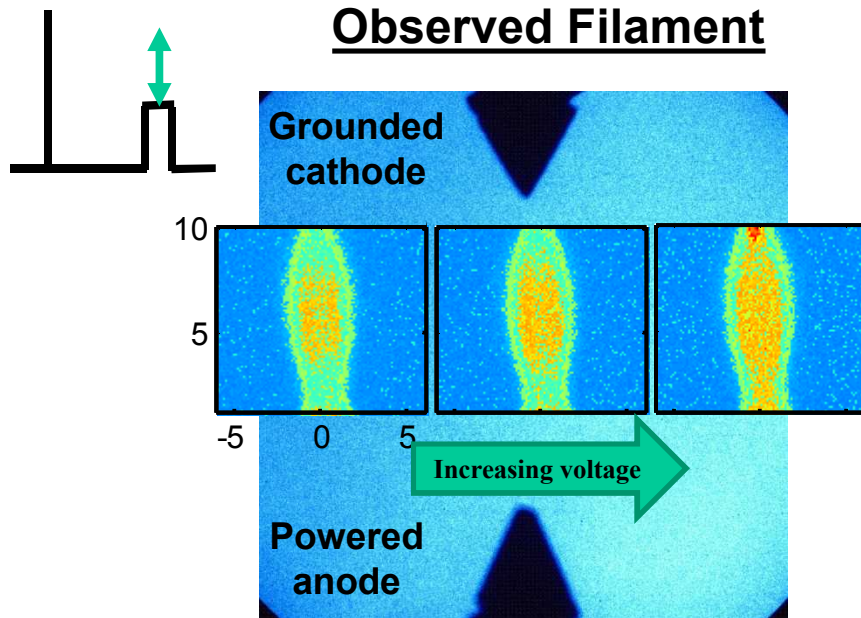
High pressure configuration



Initial efforts tried to extend earlier methods to higher pressure configuration.

DOUBLE PULSE METHOD SUCCESSFULLY DEMONSTRATED AT HIGHER PRESSURES

- Utilize observed trends to assess E/N and electron densities.
 - 400 Torr He environment.
 - Initial pulse generates filament (3 kV, 250 ns, 20 A).
 - Second, delayed pulse drives current through filament.



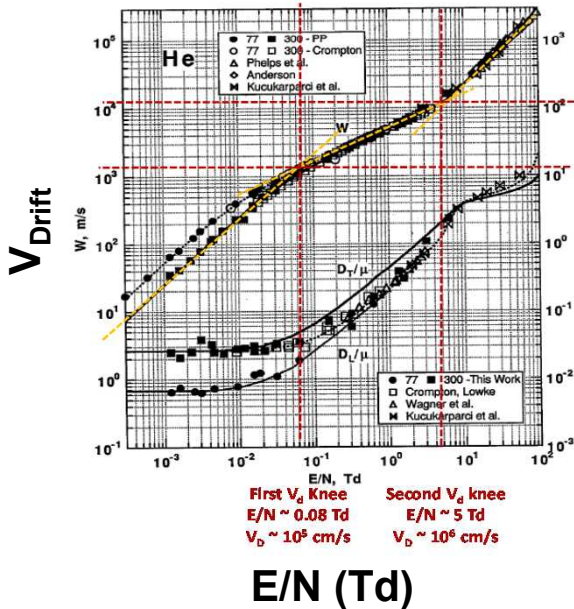
Structure of the filament does not change significantly as it is “heated”.

OBSERVATIONS BOUND PLASMA PARAMETERS IN HEATED FILAMENT

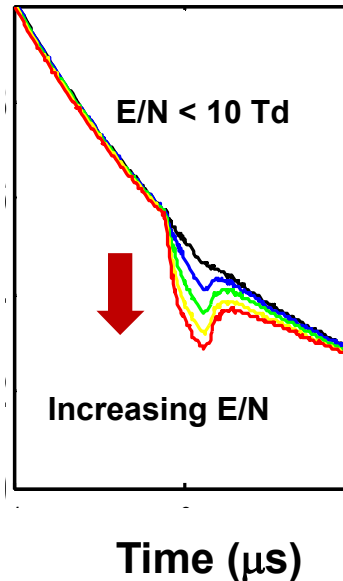
- Published drift data (Phelps) is used to bound E/N.
 - Knees in current correspond to knees in drift velocities.
 - Electron density remains roughly constant at lower E/N values.

Published drift data

J. Pack et al. JOAP, 71 (11) p5363, 1992



2^3S Densities



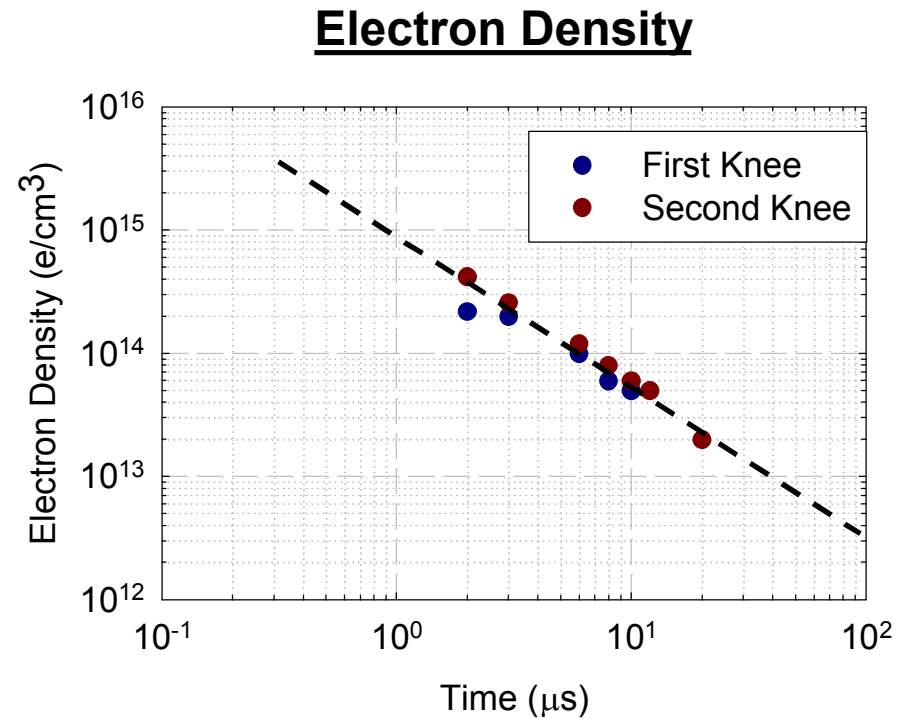
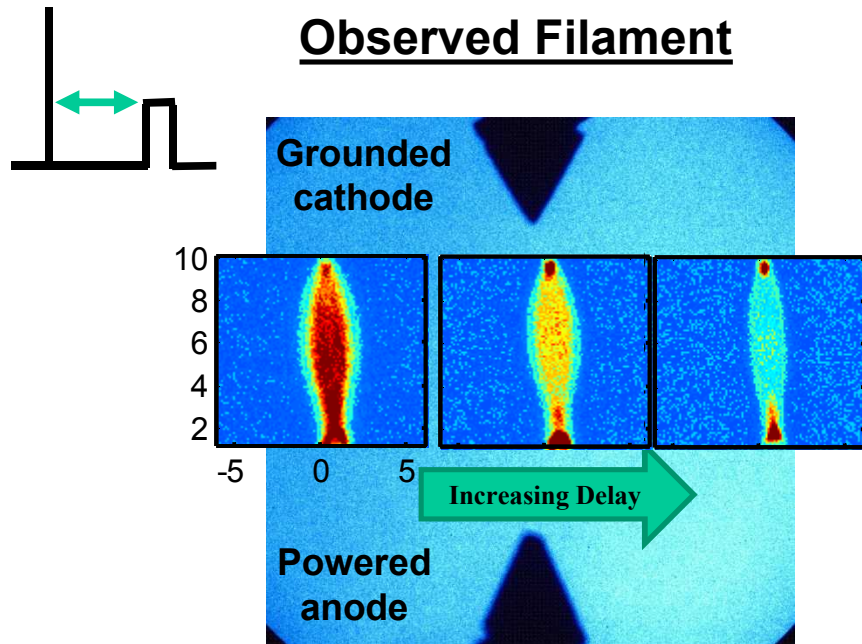
Anticipated plasma parameters

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

Observed trends yield consistent values for electron densities.

DELAY BETWEEN FIRST AND SECOND PULSE CONTROLS DENSITY

- Plasma density decays after termination of the first pulse.
 - Sweep amplitude of heating voltage to assess location of knees.

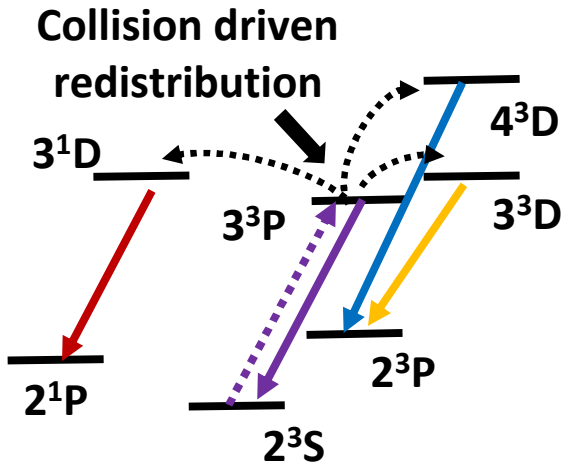


Good control of density spanning 2+ orders.

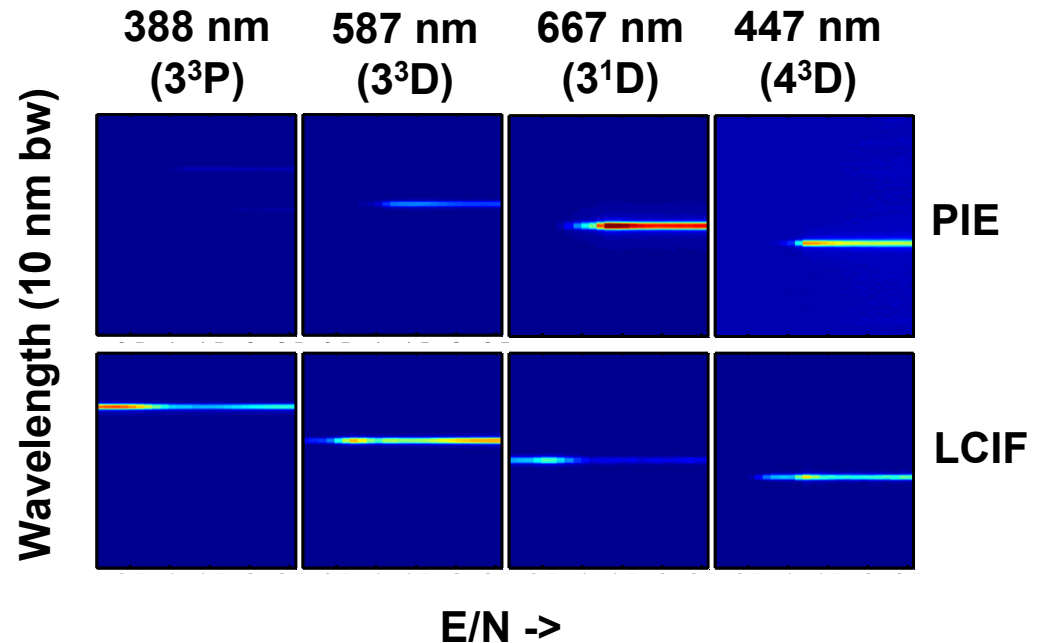
LCIF TRENDS ARE OBSERVED AT HIGHER PRESSURES

- Spectrally dispersed plasma induced emission (PIE) and LCIF and monitored as functions of E/N.
- Multiple lines from singlet and triplet manifold observed.

He levels



Observed LCIF

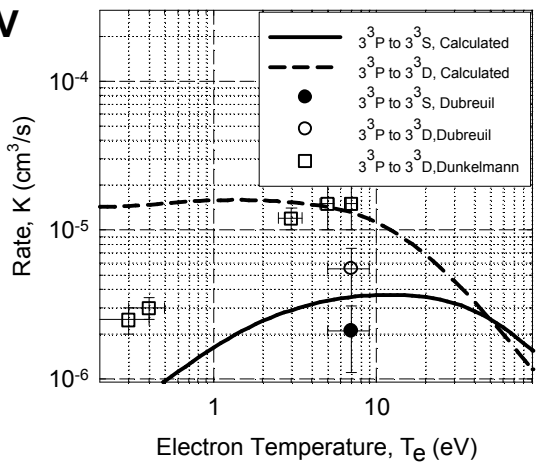
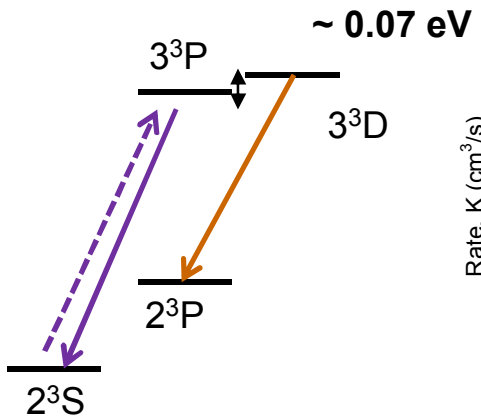


Several lines are well isolated and easily identifiable.

RATIO OF LCIF LINES ARE UTILIZED TO IDENTIFY SCALING TRENDS

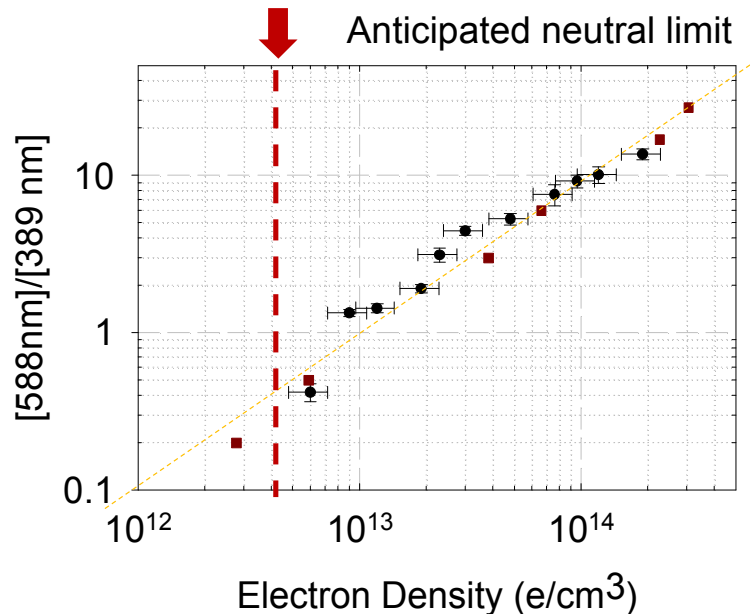
- Measurements first made as function of electron density.
 - Density varied through decay, Fixed E/N ~ 10 TD.
 - Benchmark scaling of 3^3D LCIF (weakly E/N dependent).

Pathway and Rates



Yu. Ralchenko, Atomic Data and Nuclear Data Tables **94**, 603 (2008)
 B. Dubreuil, J. Phys. B: At. Mol. Opt. Phys. **18**, 4597 (1985).

Scaling of LCIF

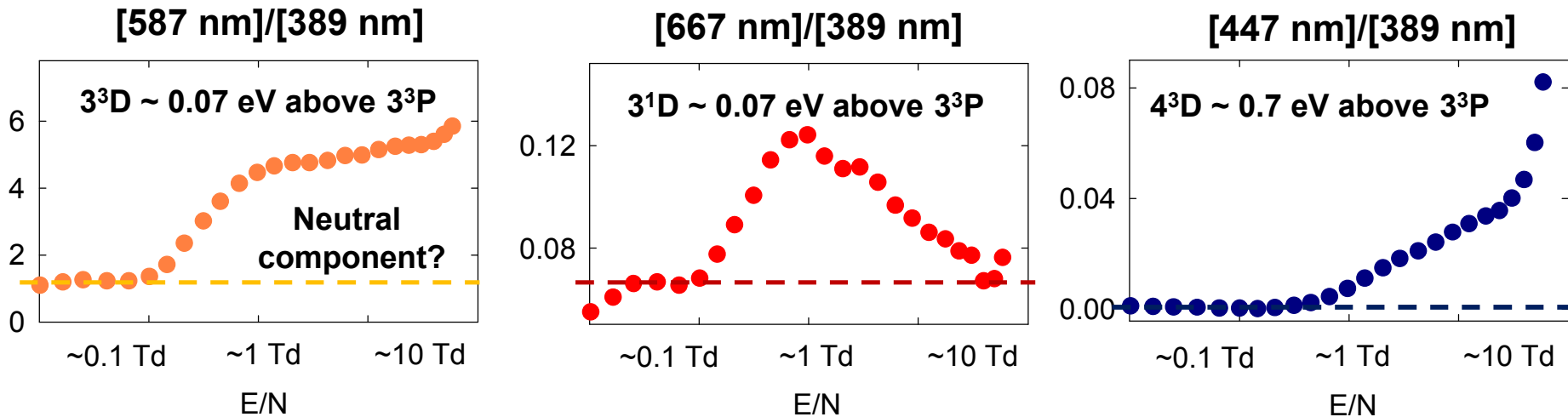


LCIF from the 3^3D demonstrates linear response with n_e .

LCIF TRENDS ARE OBTAINED AS FUNCTION OF E/N FOR FIXED ELECTRON DENSITY

- Scan heating voltage at given time after termination of first pulse.
 - Electron Density $\sim 4 \times 10^{13} \text{ e/cm}^3$.
 - Limit E/N (<10) Td to prevent ionization (Fixed n_e).

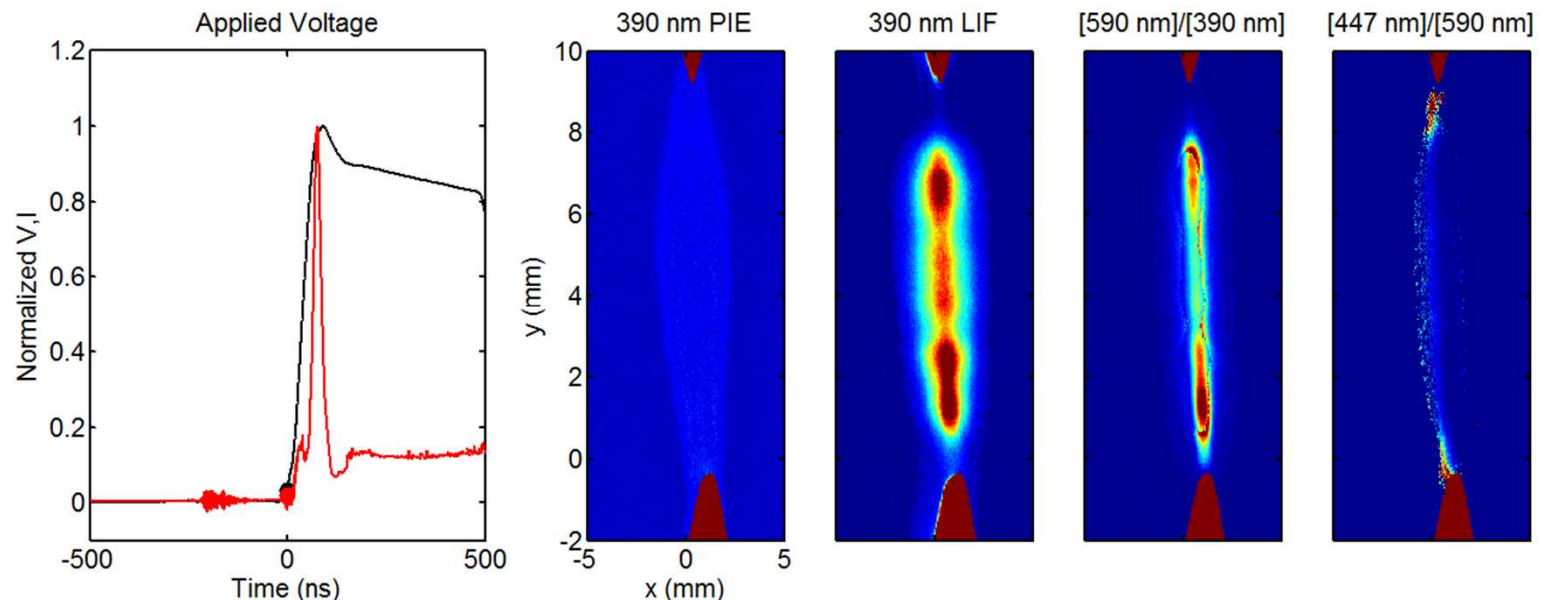
Observed LCIF trends



Each LCIF line demonstrates unique E/N dependence.

DYNAMIC EVOLUTION OF THE FILAMENT IS INTERROGATED WITH UF-LCIF

- Key strengths of LCIF is ability to provide two-dimensional images of key plasma species.
 - Examine evolution of key species as heating pulse is applied.
 - Limited beam size to suppress scattering.

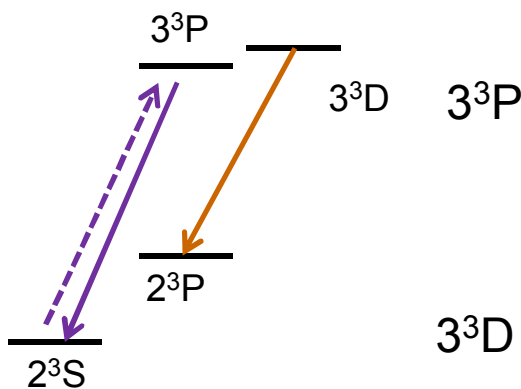


LCIF captures spatial-temporal evolution of the filament.

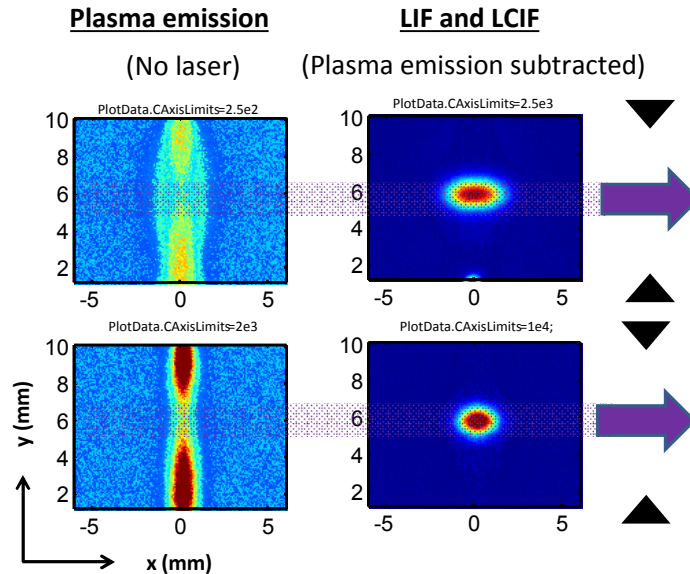
TEMPORAL EVOLUTION OF LCIF IS OBSERVED AT DIFFERENT LOCATIONS IN THE FILAMENT

- Time-stepped images are obtained of LIF and LCIF.
 - 400 Torr He Afterglow, $n_e \sim 10^{14}$ e/cm³, $n_0 \sim 10^{19}$ e/cm³.
 - Limited beam size to suppress scattering.

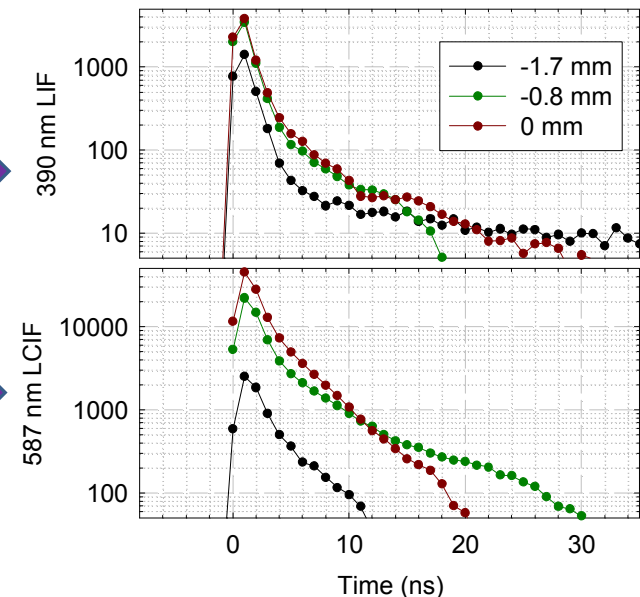
Targeted Lines



Measured L(C)IF



Time resolved L(C)IF

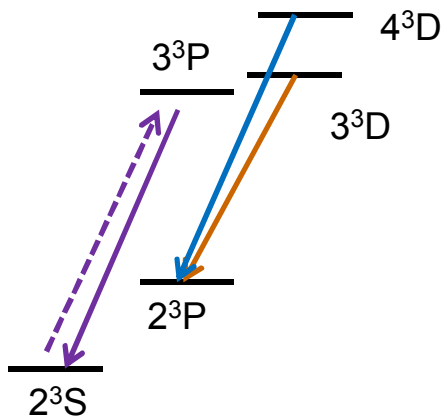


***LIF and LCIF demonstrate two characteristic decay times...
(Reason TBD)***

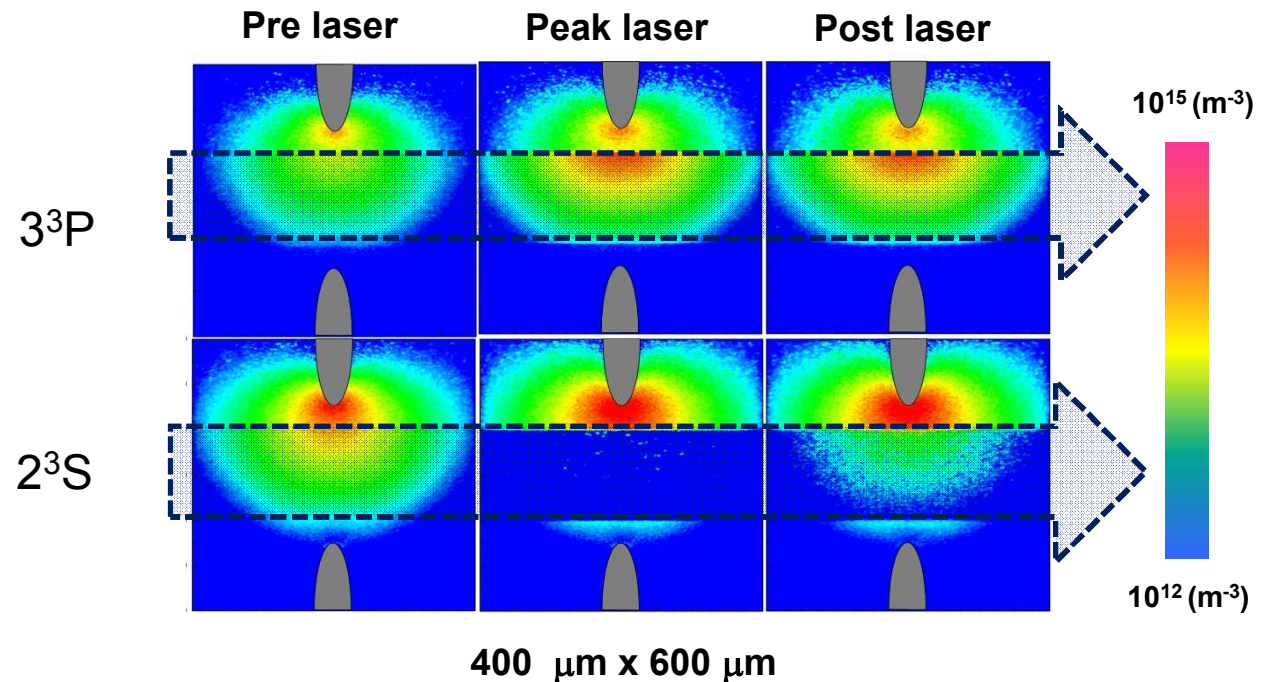
RADIATION TRANSPORT + FULLY KINETIC SIMULATIONS ARE BEING DEVELOPED AT SNL

- Simulation of plasma response to 100 ps laser excitation.
 - ALEPH – 3D fully kinetic PIC code (M. Hopkins, Poster)

Targeted Lines



Simulated LCIF (A. Fierro)

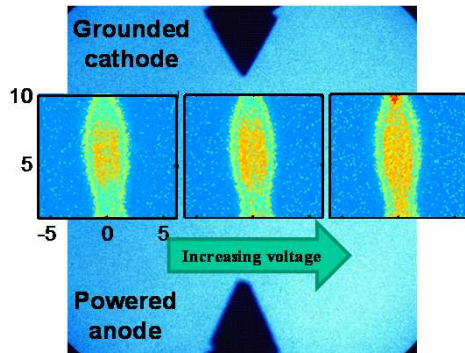


Observations are being used to benchmark preliminary results

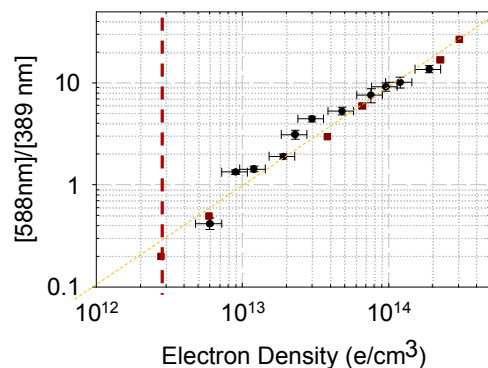
SUMMARY OF ULTRAFAST LASER-COLLISION INDUCED FLUORESCENCE STUDY

- First steps focused on building on earlier methods utilized for calibrating LCIF.
 - Double pulse method for generating tuned n_e , T_e .
- Preliminary results look promising.
 - Favorable scaling of detectable LCIF.
- A few hiccups that warrant further understanding.
 - Temporal evolution and radiation transport.

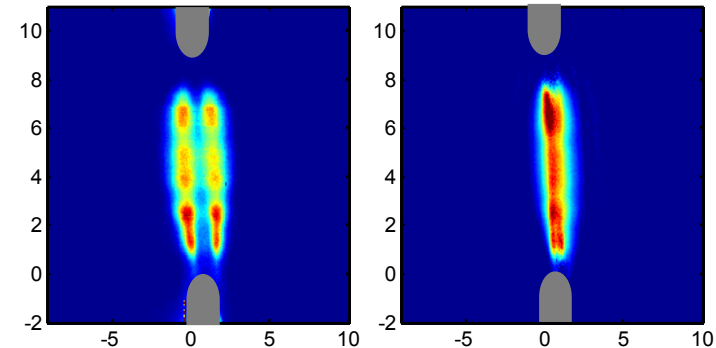
Setup



Scaling



Implementation



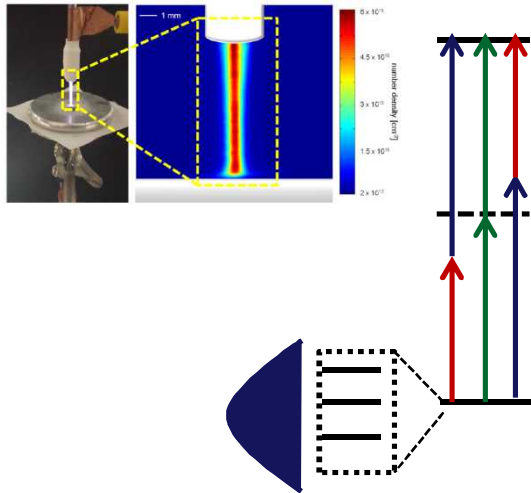
Optimistic about utility of UF-LCIF for high pressure plasma interrogation!

FUTURE DIRECTIONS FOR UF PLASMA DIAGNOSTIC DEVELOPMENT AND IMPLEMENTATION

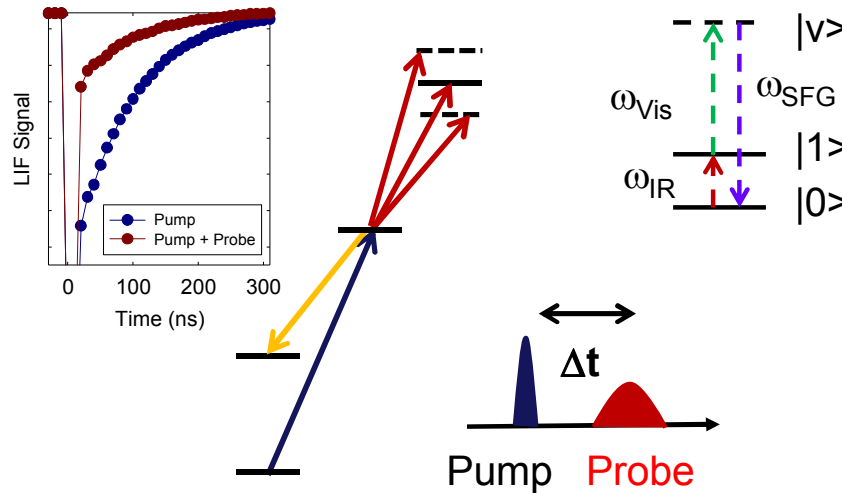
- Ultrafast, high intensity lasers open up several venues for interrogating challenging plasma environments.
 - Efficient planar two-photon LIF of gas-phase species.
 - Pump-probe spectroscopy of fast phenomenon.
 - Surface spectroscopy on molecular species.

Short-pulse two photon

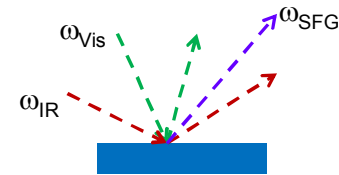
J. Schmidt, PSST 24,032004 (2015)



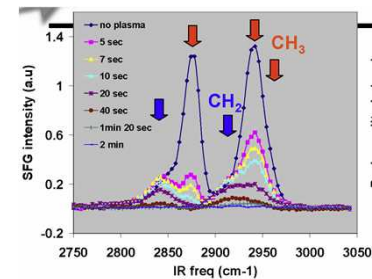
Ultrafast pump-probe



Surface Spectroscopy



D. Farrow, SNL



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,...).**
- **Short-pulse lasers show promise for other measurements of interest.**
 - **Atomic species, collision rates, electric fields, interfaces.....**

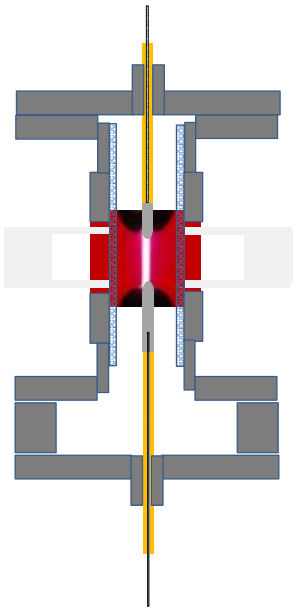
Thank you for attention and continued support!

AUXILARY SLIDES

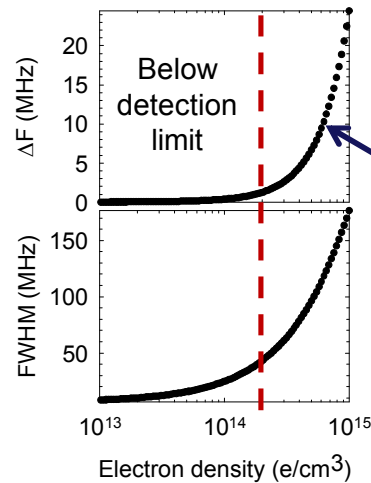
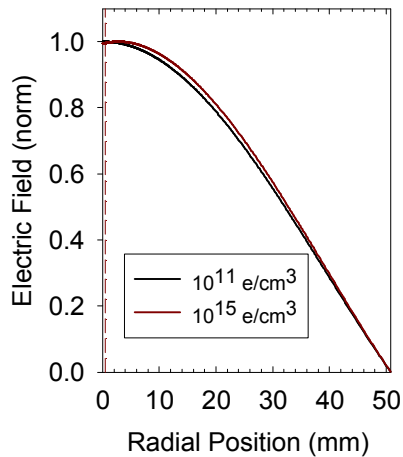
INITIAL ATTEMPTS EMPLOYED RESONANT CAVITY TO MEASURE DENSITIES

- Used in house EM solver to predict the response of the cavity to the plasma filament
 - Despite high pressures, detecting shifts and broadening are manageable

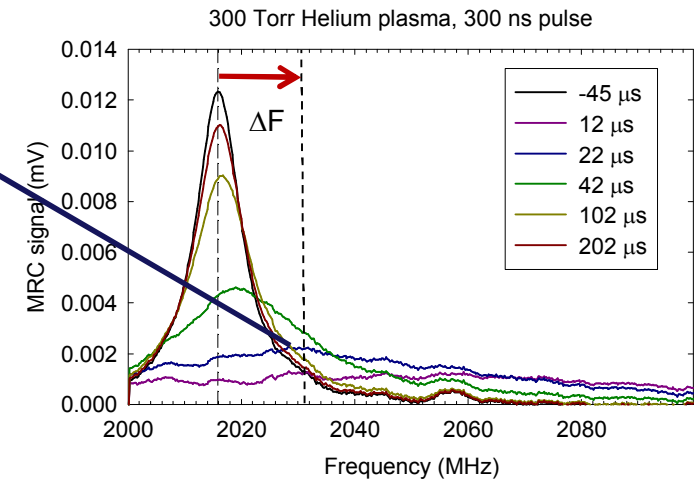
Setup



EM Solutions



Measured response

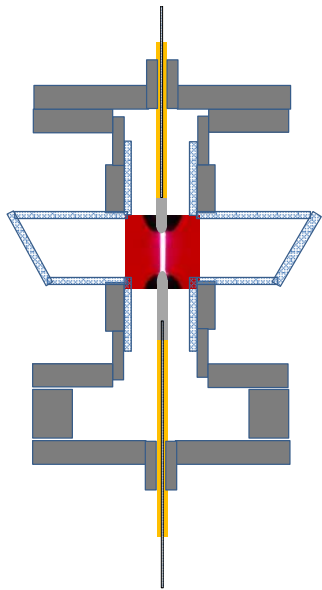


Resonant cavity can work but the challenge is perturbation of electrode and limited dynamic range

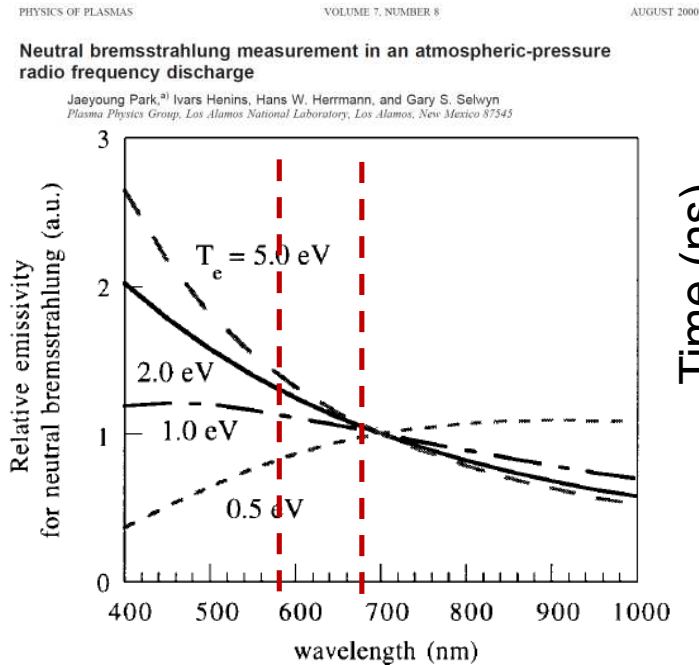
CONTINUUM EMISSION GENERATED IN HIGH PRESSURE HELIUM PLASMA

- Continuum emission generated by electron-neutral bremsstrahlung
 - Potentially useful for assessing T_e (shape) and n_e (Intensity)

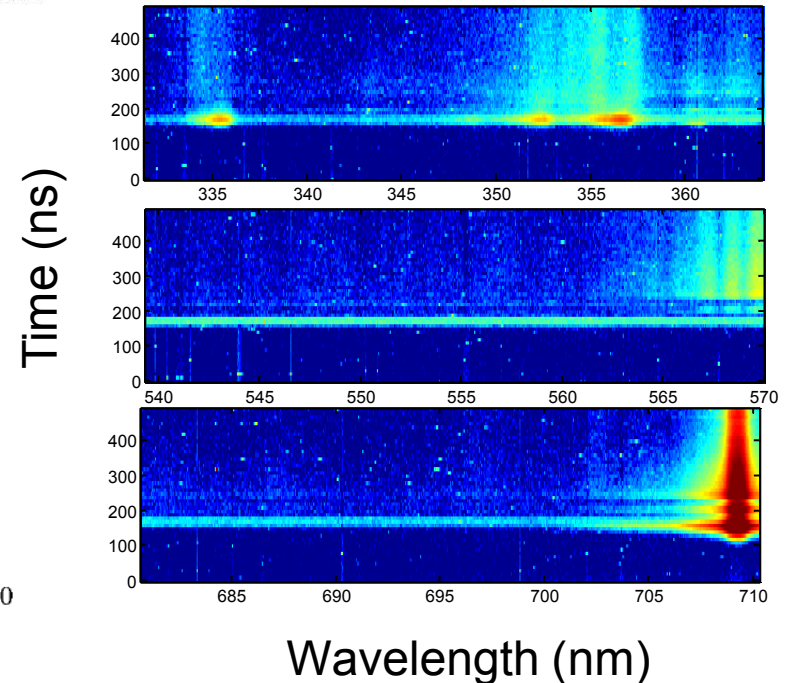
Setup



Anticipated scaling



Observed Emission

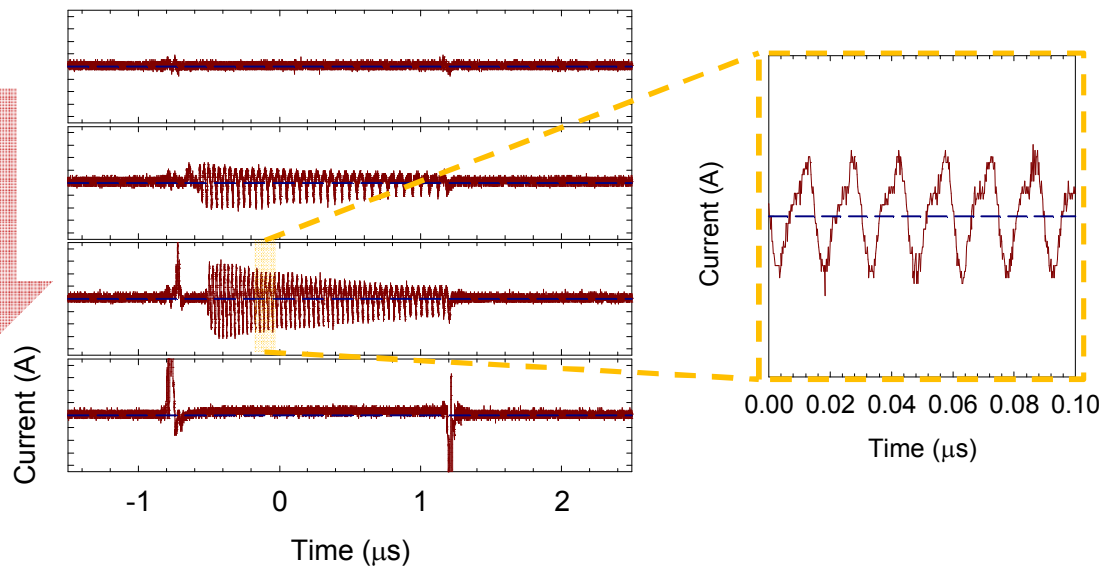


Broadband emission is tempting target to quantify 'Te' but spectral "contamination" limits use

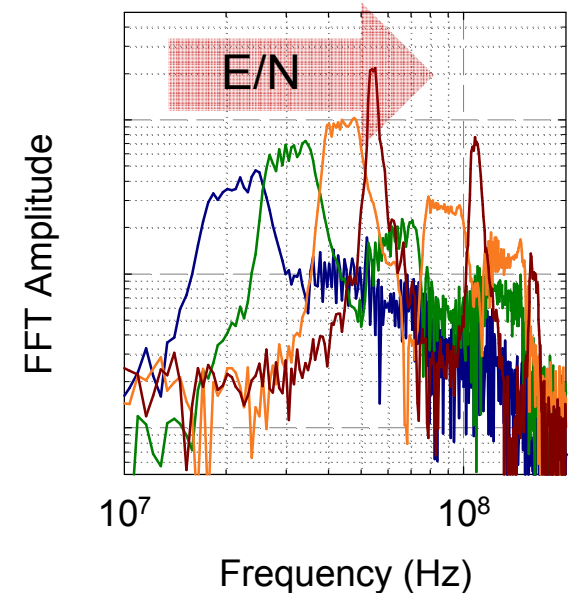
HIGH FREQUENCY OSCILLATIONS OBSERVED AT LOW E/N VALUES

- Instabilities observed in cathode current waveforms
 - Appear at low E/N values (~ 0.1 Td)
 - Does not seem to be indicative of change in structure
 - Have sporadic nature... not always repeatable

Cathode current



FFT of Current

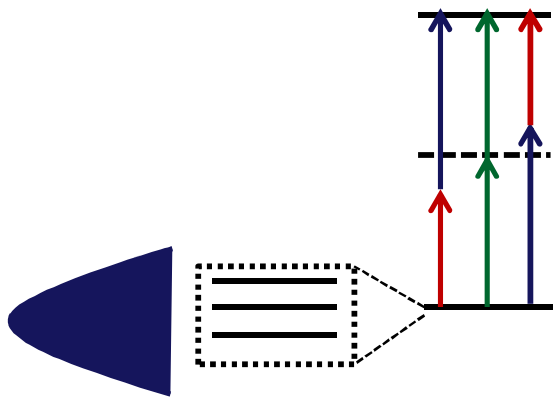


Characteristic frequency increases and sharpens with E/N.... then turns off.

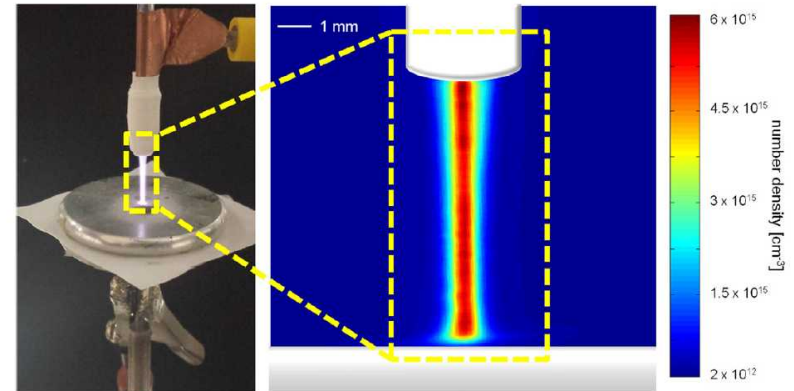
TWO-PHOTON EXCITATION WITH INTENSE UF LASER PULSES

- Ultrafast lasers offer distinct advantages for two-photon excitation
 - High intensities (1 mJ @ 100 fs = 10^{10} W ; 1 mJ @ 10 ns = 10^5 W)
 - Broadband (10's nm) excitation (Overlap + multiple transitions)
 - Coherent sum over two-photon excitation
 - Reduced photolytic production of atomic species

Short-pulse two photon



Planar UF two-photon LIF



J. Schmidt, PSST 24,032004 (2015)

Broad range of relevant species are accessible (O, N, H)

MASTER SLIDE TEMPLATE FOR ADDITIONAL CONTENT

- Text should be Arial 20 pt (18 pt if you need more text) with bullets from the symbol list. Line spacing for major bullets should be 0 points before, 6 pts after.
 - This is a sub-bullet line
 - This is another sub-bullet line
 - Last line should have spacing 6 pts after.
- Insert figures as TIFF, JPEG or GIF files.
 - Animations should be GIF files that are self contained (that is not external files)
 - I can convert AVI and other formats to GIF
- Another major bullet
 - Another sub-bullet.