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DEVELOPMENT OF LASER-COLLISION INDUCED FLUORESCENCE FOR ATMOSPHERIC PRESSURE PLASMA GENERATED IN HELIUM ATMOSPHERES

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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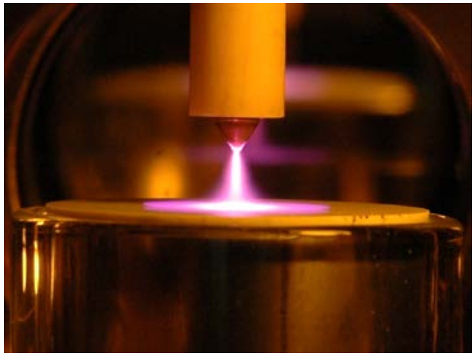
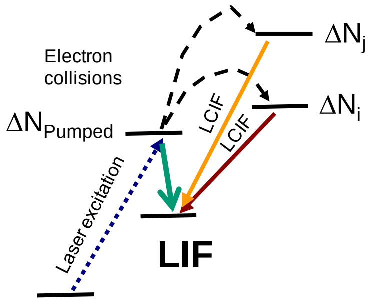
INTRODUCTION AND MOTIVATION: CHALLENGES AND OBJECTIVES

- **High pressure (~ 1 ATM and beyond) plasma is challenging environment.**
  - **Higher densities.**
  - **Chemically complex environments**
  - **Smaller length scales.**
  - **Shorter lifetimes.**
  - **Optically thicker environments.**
- **Investigate diagnostics methods to access this challenging environment**
  - **Extend laser-collision induced fluorescence (LCIF)**
  - **Examine suitability of ultrafast-short pulse lasers for use**





Overall goal is to match potential opportunities offered by short-pulse lasers to challenges offered by high pressure plasmas.

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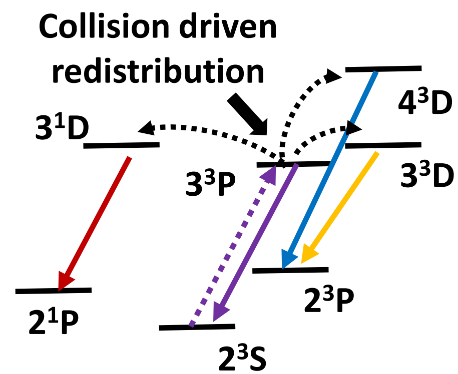
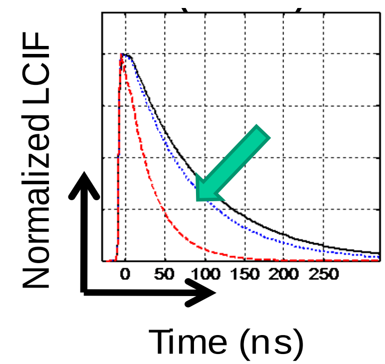
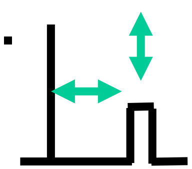
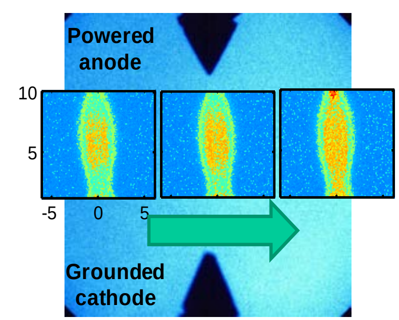
STRATIGIES USED TO DEVELOP LCIF AT HIGHER PRESSURES

- **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 << decay times to simplify interpretation.**
  - **Stepping stone for other methods.**
- **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.

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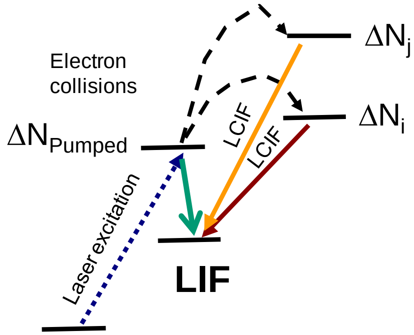
OVERVIEW OF CHALLENGS OF EXTENDING LCIF TO HIGH PRESSURE

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



"Electron mixing" ***"Neutral mixing"*** ***"Photon mixing"***

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

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

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

Simplifications likely not to be so forthcoming at higher pressures

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

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

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 ~ 10^{13} e/cm^3 at 1000 Torr

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

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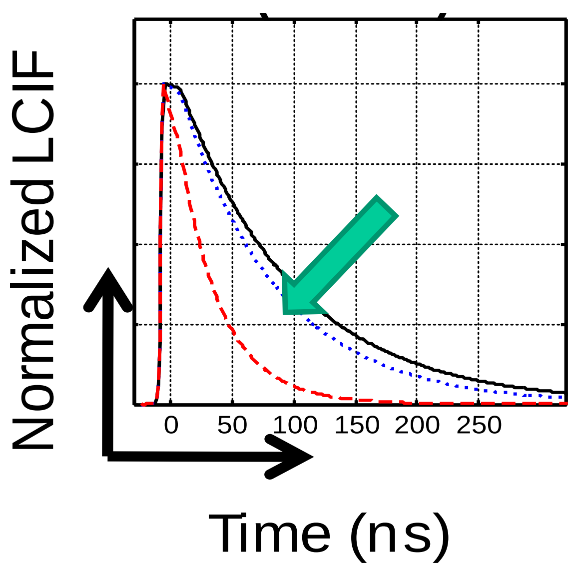
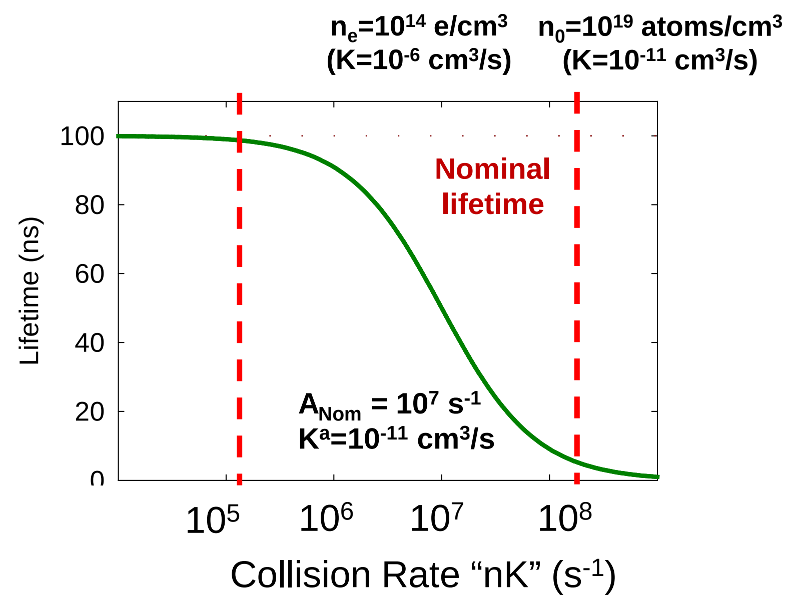
LIFETIMES OF EXCITED STATES BECOME VERY SHORT AT HIGHER ELECTRON DENSITIES

- **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.

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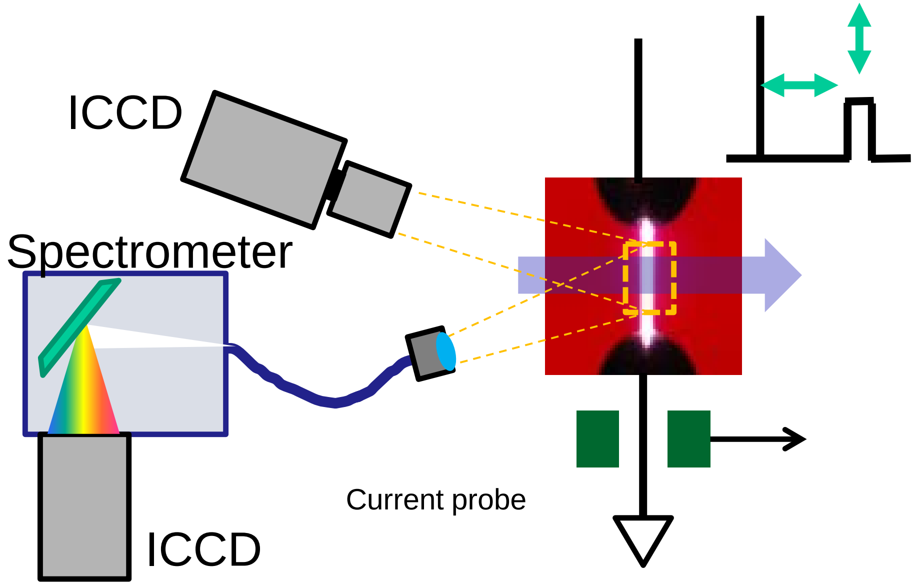
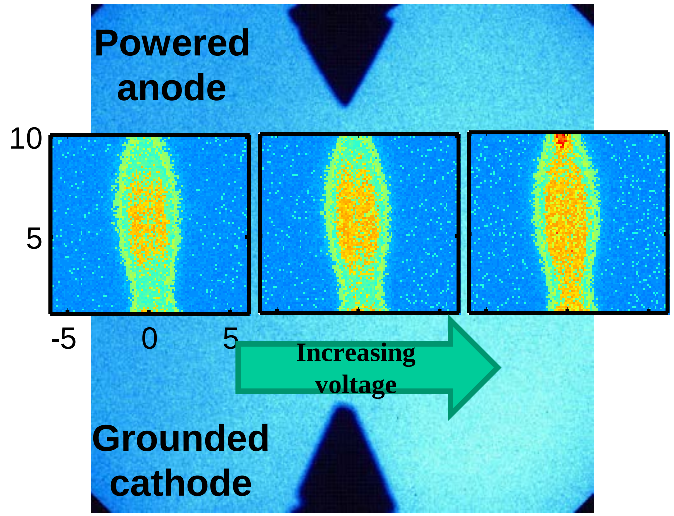
KEY CHALLENGE: GENERATING AND MANIPULATING WELL CHARACTERIZED PLASMA

- **Previous experience in developing LCIF indicated that setup is key.**
  - **Plasma generation in 640 Torr He.**
  - **Double pulse method to separate generation and interrogation.**
  - **Spectrometer to identify, camera to image.**

High pressure configuration



Observed Filament



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

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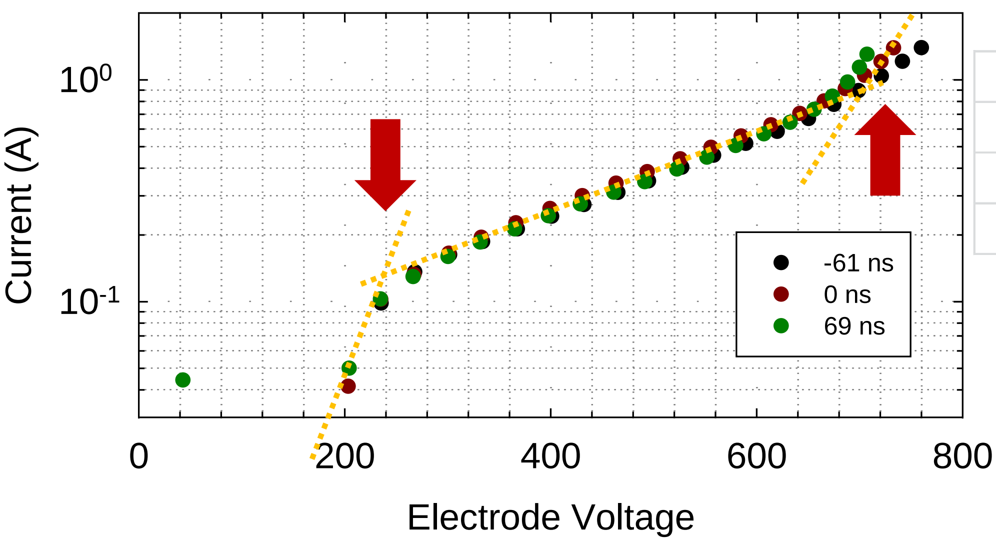
UTILIZE CURRENT-VOLTAGE TRENDS TO BOUND PLASMA PARAMETERS

- **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

V	I	J	E/N	Vd	ne
V	A	A/cm2	Td	cm/s	e/cm3
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

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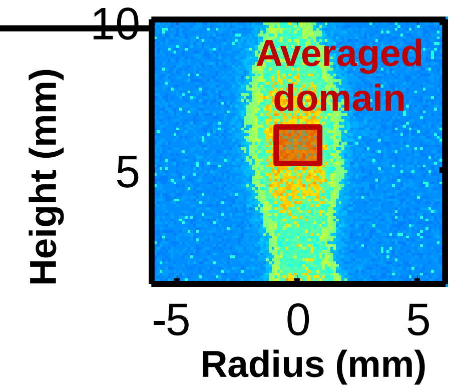
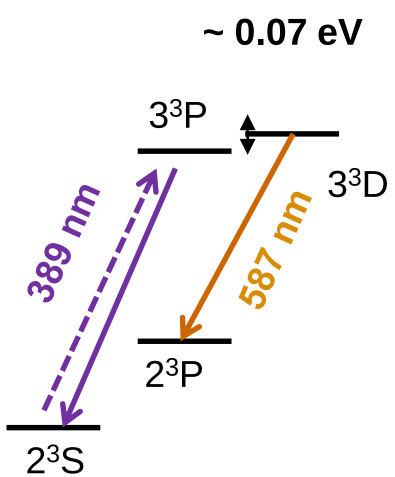
J. Pack et al. JOAP, 71 (11) p5363, 1992

RATIO OF LCIF LINES ARE UTILIZED TO IDENTIFY SCALING TRENDS

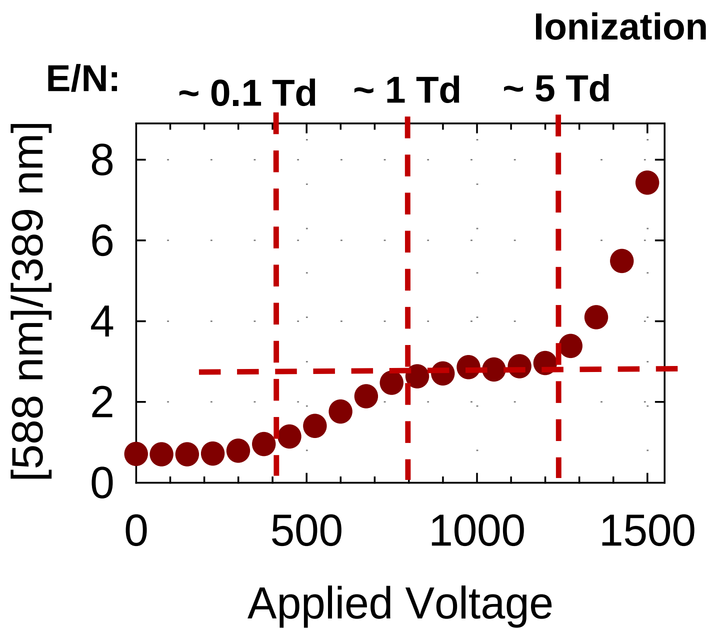
- **Benchmark scaling of 3^3D LCIF.**
  - **Averaged from central region of the discharge.**



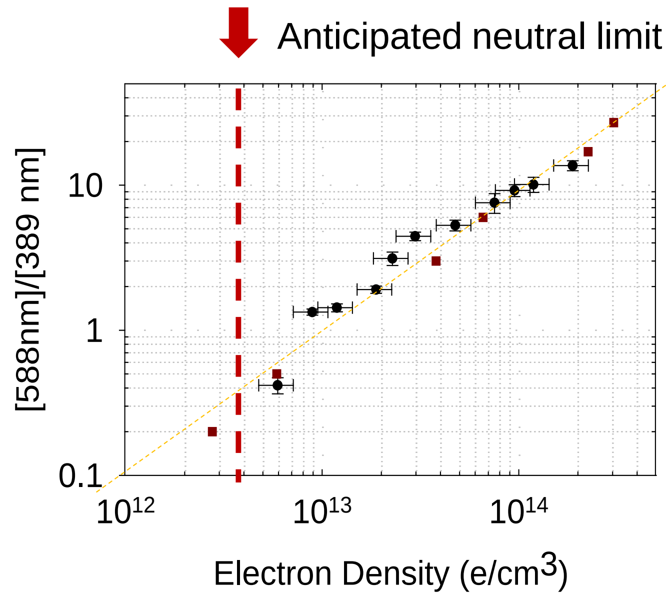
Pathway



Scaling of LCIF



$n_e \sim 2 \times 10^{13}$ e/cm^3



E/N ~ 5 Td

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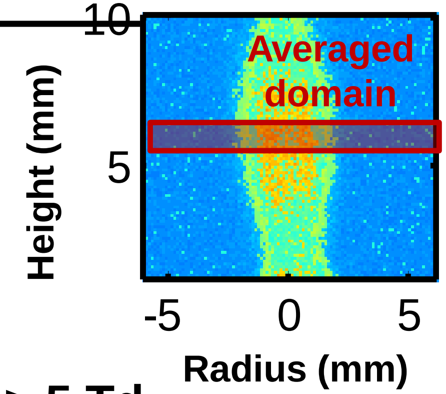
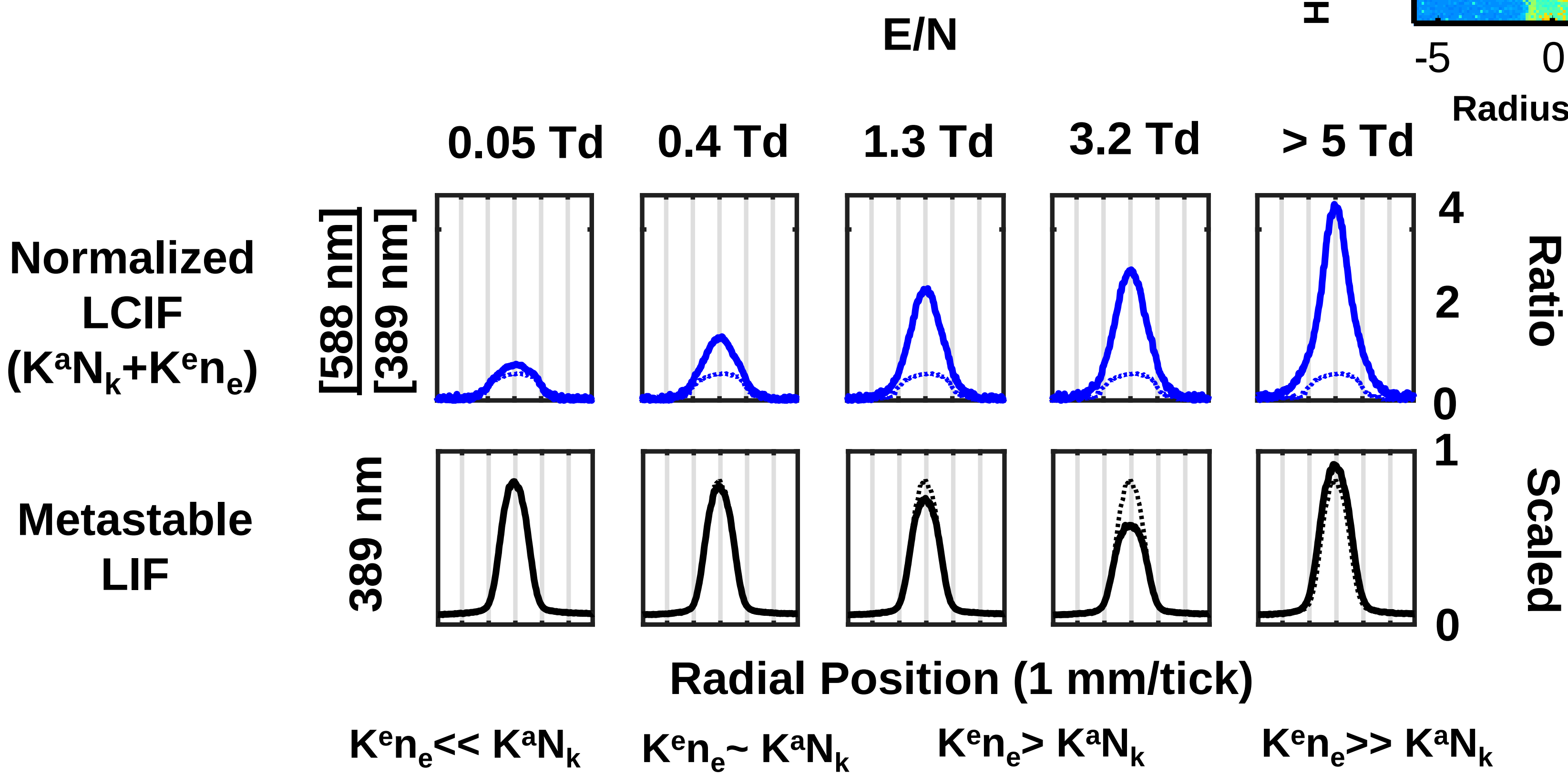
LCIF provides strong signals and easily detectable over broad range of conditions

DEMONSTRATION OF SPATIAL RESOLUTION PROVIDED BY LCIF

- **Radial structure observed for various E/N.**
  - **Initial peak electron density of $2x10^{13}$ e/cm^3**





Observations consistent with initial assertions made

Onset of ionization

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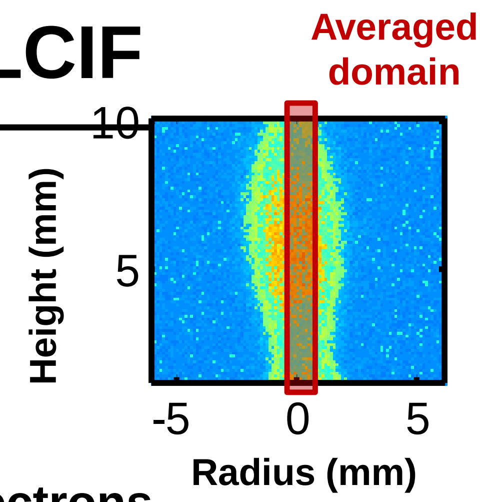
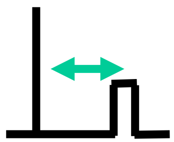
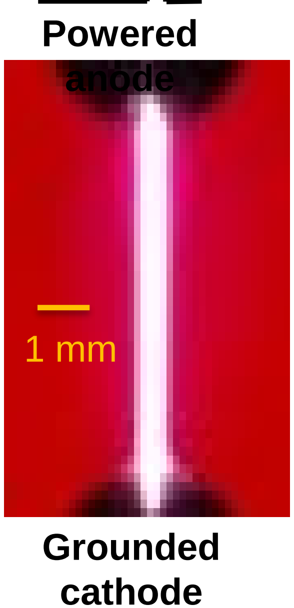
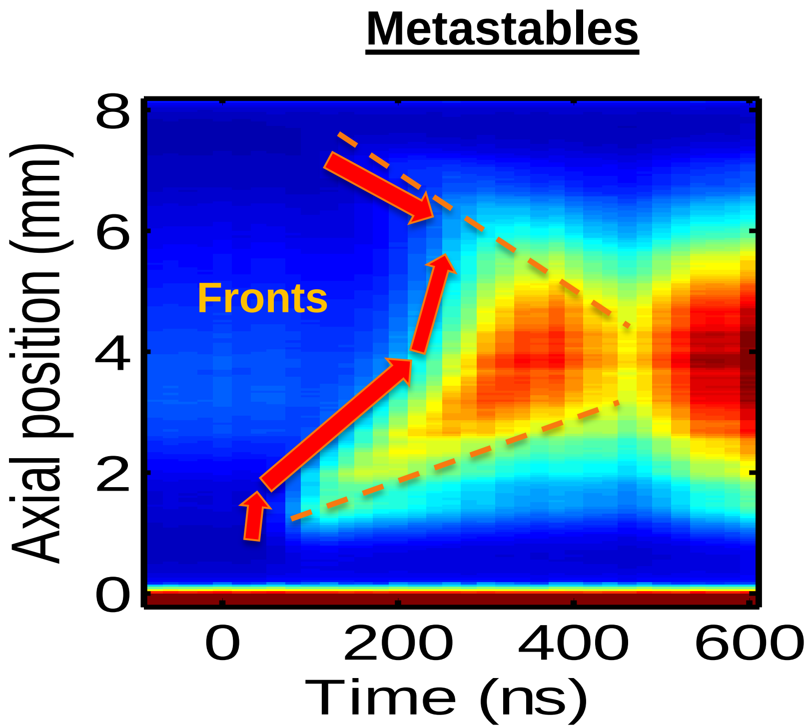
DEMONSTRATION OF SPATIAL RESOLUTION PROVIDED BY LCIF

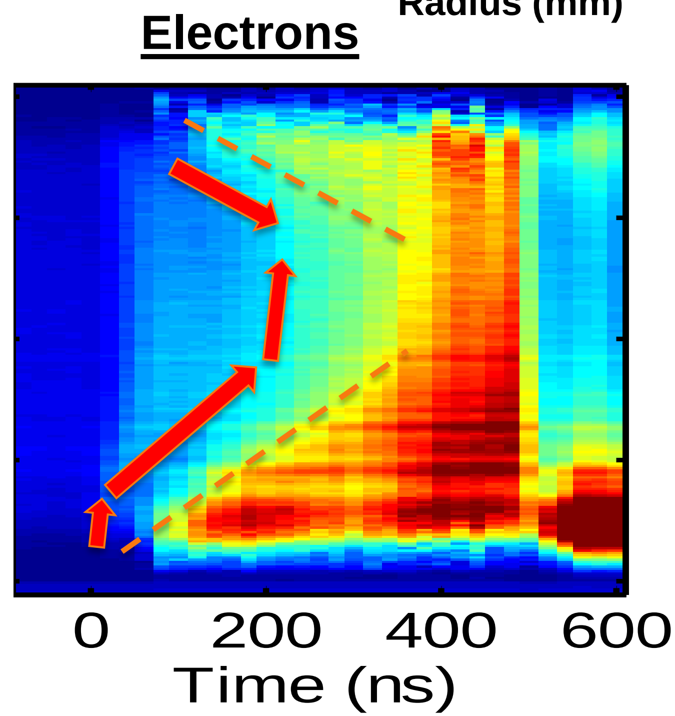
- **Streak-like images of plasma initiation.**
  - **Radial profiles at center of discharge.**











LCIF captures spatial and temporal evolution of plasma formation.

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 and lifetimes..**

Thank you for attention!

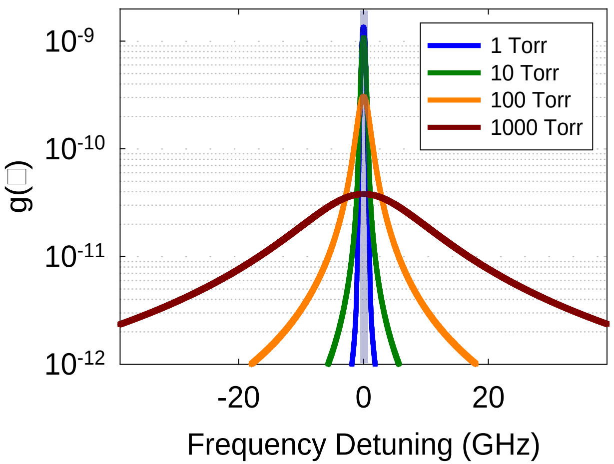
This work was supported by the Department of Energy Office of Fusion Energy Science
Contracts DE-SC0001939 and DE-AC04-94SL85000

AUXILARY SLIDES

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)

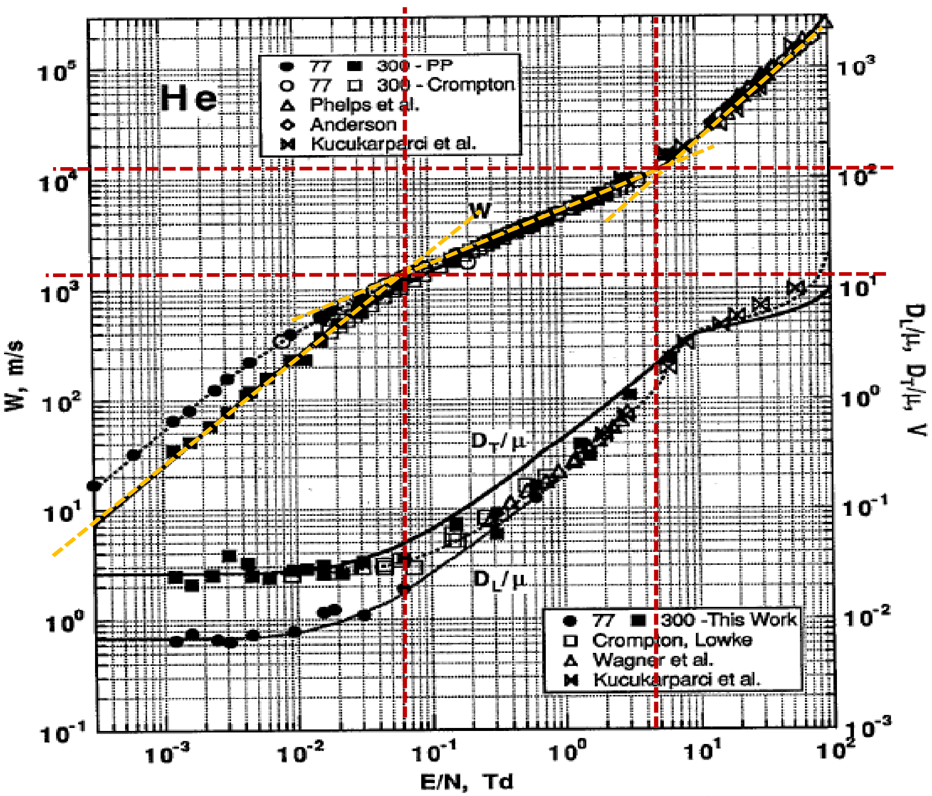
Short pulse enables access to all of the pressure-broadened line.

UTILIZE CURRENT-VOLTAGE TRENDS TO BOUND PLASMA PARAMETERS

Published drift data

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

V_{Drift}



First V_d Knee
E/N ~ 0.08 Td
V_D ~ 10^5 cm/s

Second V_d knee
E/N ~ 5 Td
V_D ~ 10^6 cm/s

E/N (Td)

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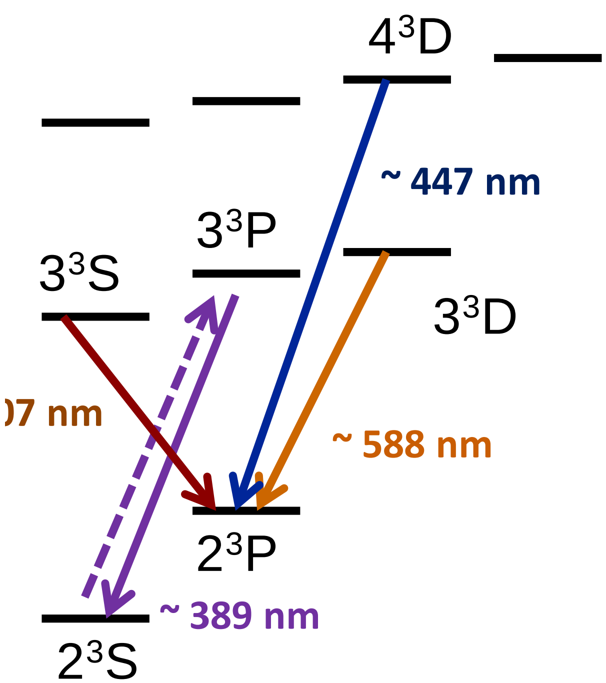
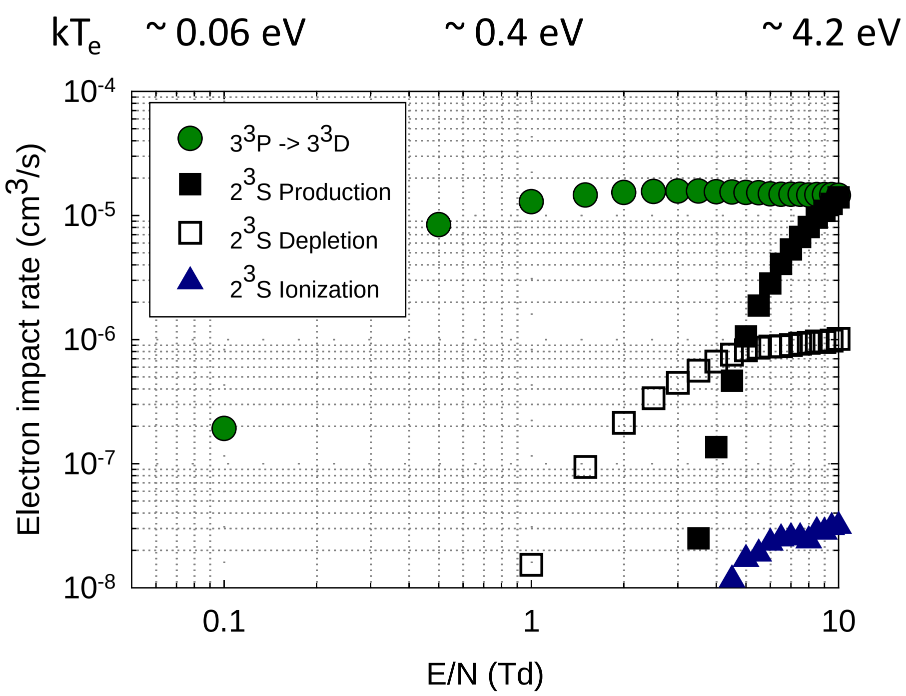
RATIO OF LCIF LINES ARE UTILIZED TO IDENTIFY SCALING TRENDS

Pathway and Rates





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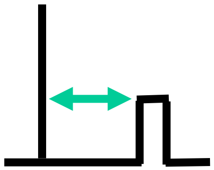
LCIF from the 3^3D demonstrates linear response with n_e.

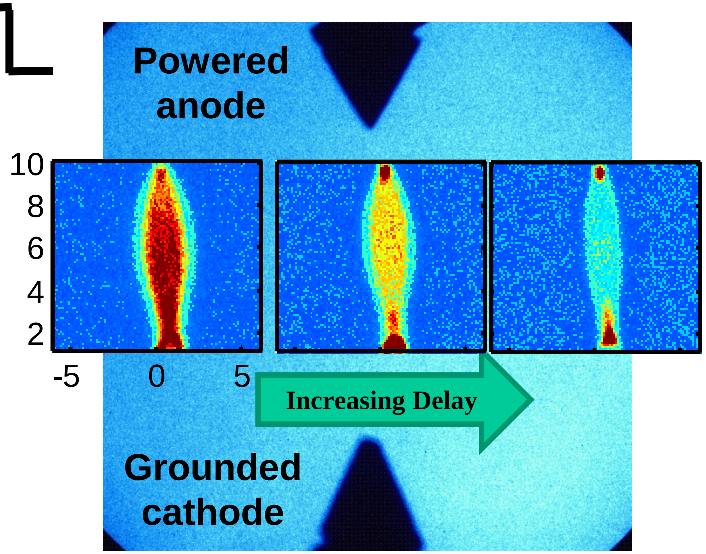
Yu. Ralchenko, Atomic Data and Nuclear Data Tables **94**, 603 (2008)
B. Dubreuil, J. Phys. B: At. Mol. Opt. Phys. **18**, 4597 (1985).

DELAY BETWEEN FIRST AND SECOND PULSE CONTROLS ELECTRON DENSITY

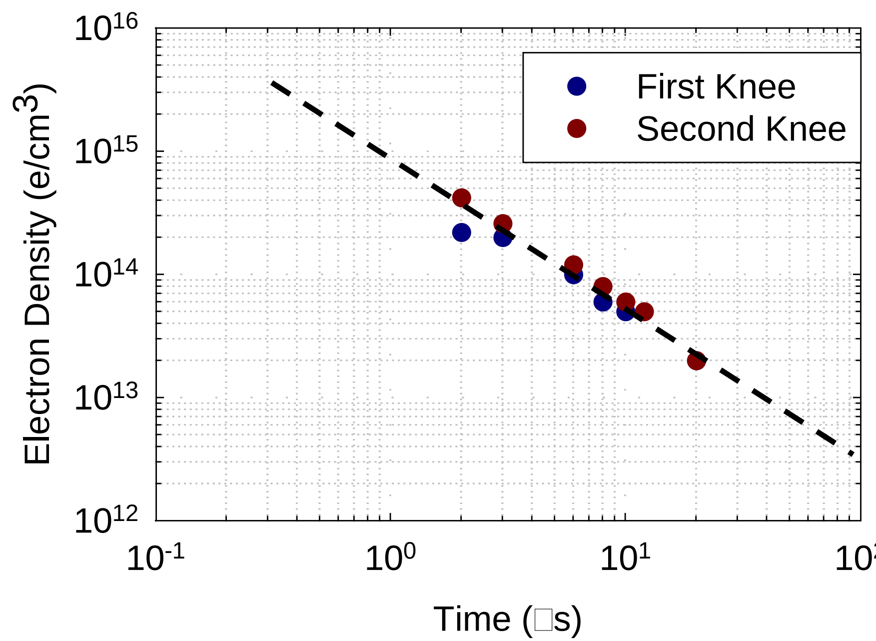
- **Plasma density decays after termination of the first pulse.**
  - **Sweep amplitude of heating pulse to assess location of knees.**



Observed Filament



Electron Density



Good control of density spanning 2+ orders.