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

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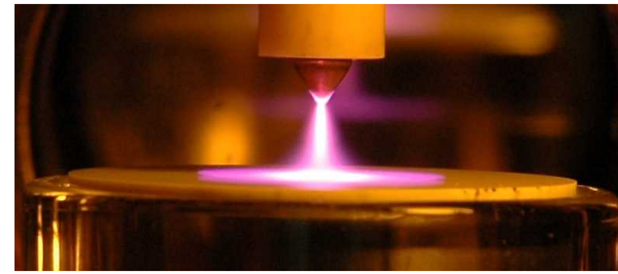
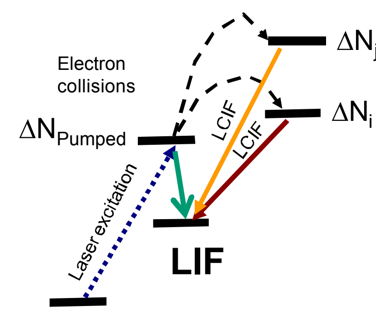
INTRODUCTION AND MOTIVATION: CHALLENGES AND OBJECTIVES

- **High pressure (~ 1 ATM and beyond) plasma is challenging environment.**
  - **Higher densities.**
  - **Chemically complex environments**
  - **Smaller length scales.**
  - **Shorter lifetimes.**



- **Investigate diagnostics methods to access this challenging environment**
  - **Extend laser-collision induced fluorescence (LCIF) to atmospheric pressure helium discharge.**
  - **Examine suitability of ultrafast-short pulse lasers for use to these environments.**



Outline challenges and demonstrate implementation of LCIF to highly collisional environments.

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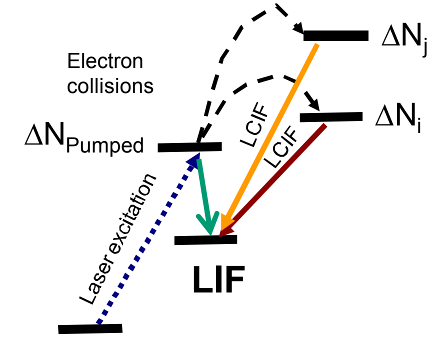
OVERVIEW OF CHALLENGS OF EXTENDING LCIF TO HIGH PRESSURE

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



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

$$\frac{dN_j}{dt} = \left[\sum_{i\neq j} K^e_{ij} N_i - \sum_{i\neq j} K^e_{ji} N_j\right] n_e + \sum_k \left[\sum_{i\neq j} K^a_{ikj} N_i - \sum_{i\neq j} K^a_{jki} N_j\right] N_k + \left[\sum_{i>j} A_{ij} N_i - \sum_{i<j} A^j_{ji} N_j\right]$$

$$\Delta N_j \sim K^e_{ij} n_e \times \Delta N_i \times \Delta t$$

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

Simplifications likely not to be so forthcoming at higher pressures

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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

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

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

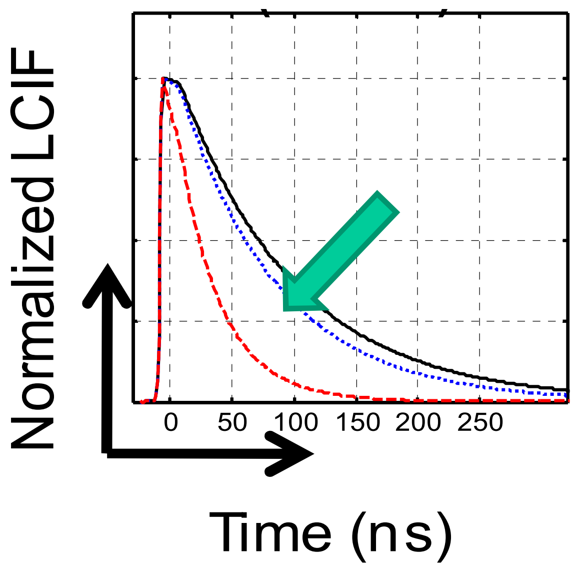
Limit ~ 10^{13} e/cm^3 at 1000 Torr

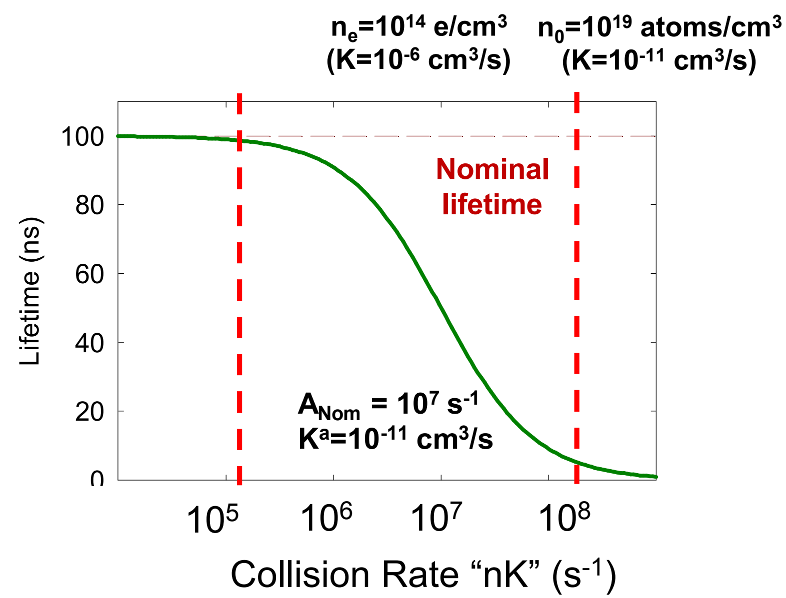
Neutrals are anticipated place limits on lower bound of electron detection.

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.

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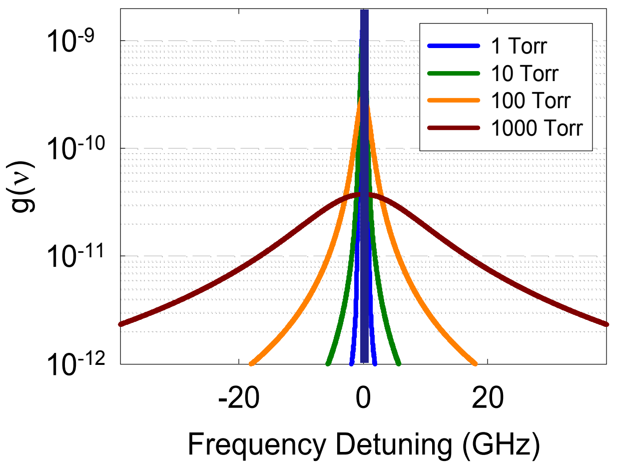
ULTRASHORT-PULSE LASER IS USED FOR INITIATION LCIF EVENT

- 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)

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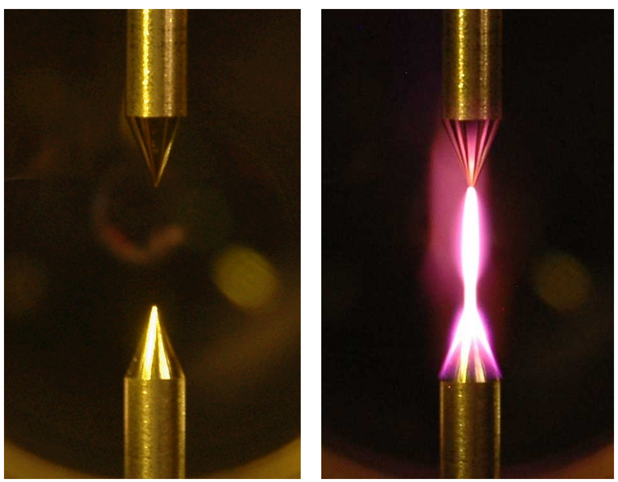
Short pulse enables access to all of the pressure-broadened line.

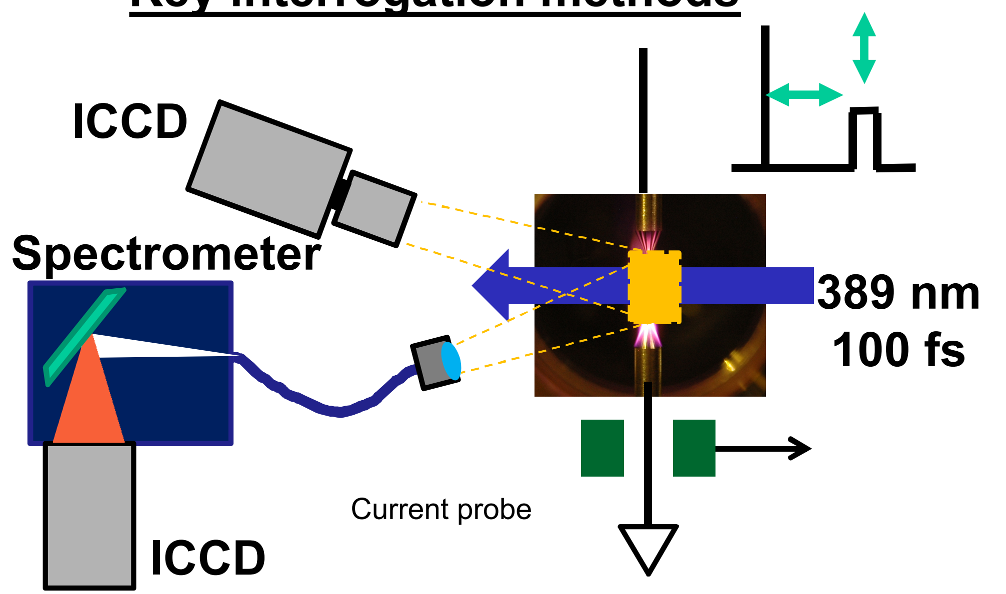
KEY CHALLENGE: GENERATING AND MANIPULATING WELL CHARACTERIZED PLASMA

- **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.

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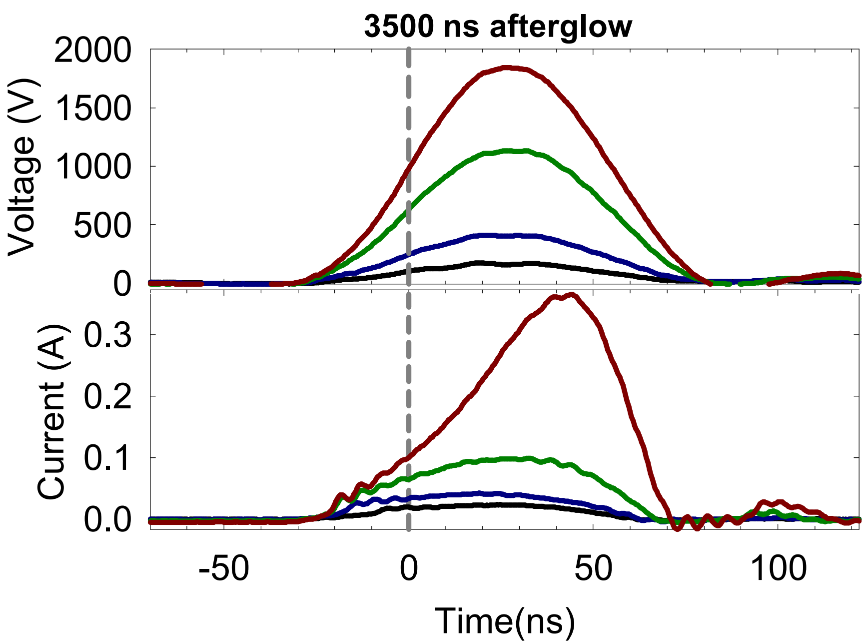
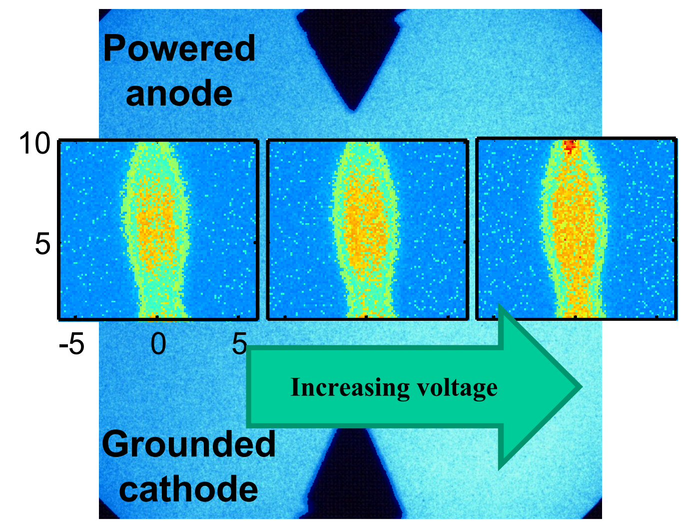
DOUBLE PULSE PLASMA SEPERATES GENERATION FROM MANIPULATION

- **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



Observed Filament



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

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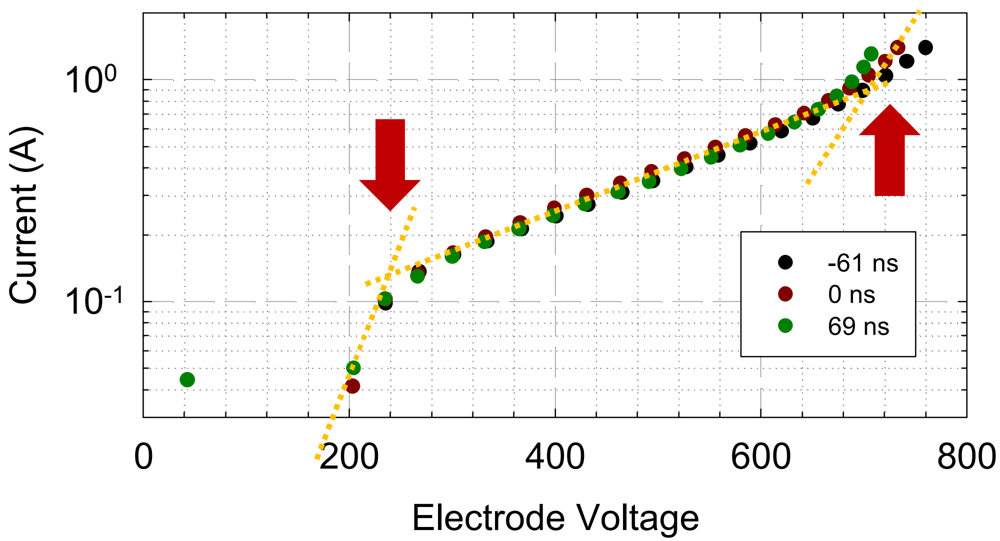
UTILIZE CURRENT-VOLTAGE TRENDS TO BOUND PLASMA PARAMETERS

- **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

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

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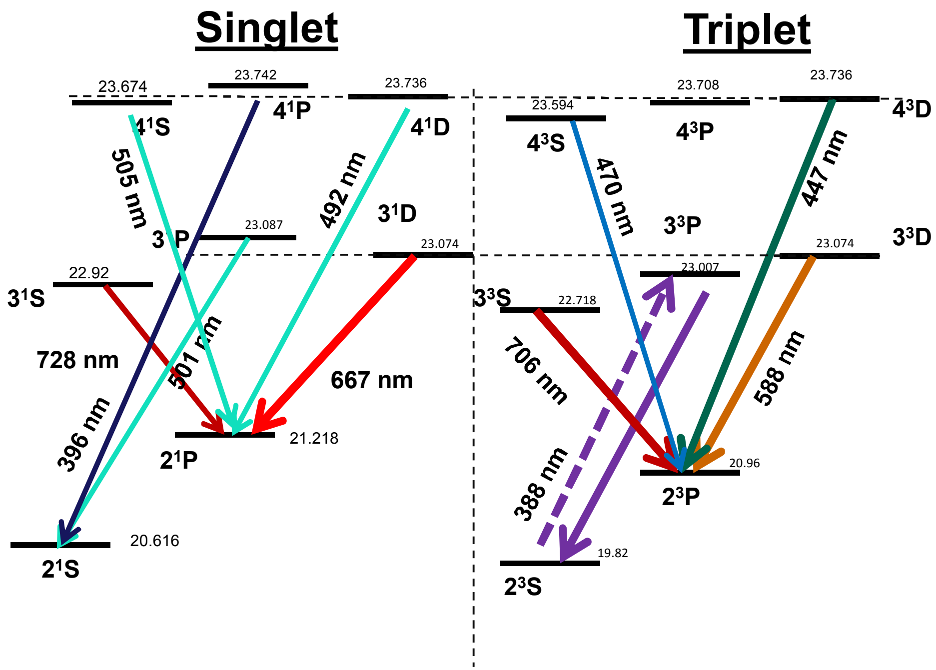
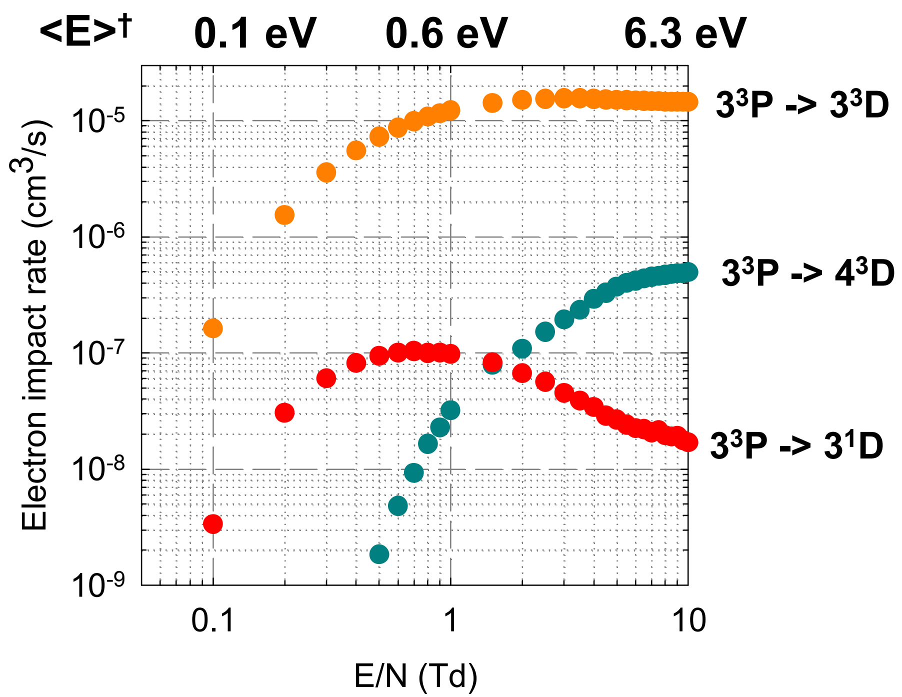
J. Pack et al. JOAP, 71 (11) p5363, 1992

LCIF EVOLVES THROUGH SEVERAL SPECTROSCOPIC PATHWAYS

- **Transitions from the laser excited 3^3P state have different E/N scaling**
  - **Capitalize on unique characteristics to assess <E> and E/N.**

Pathway and Rates





Identify pathways that yield good signals and are "easy to" evaluate.

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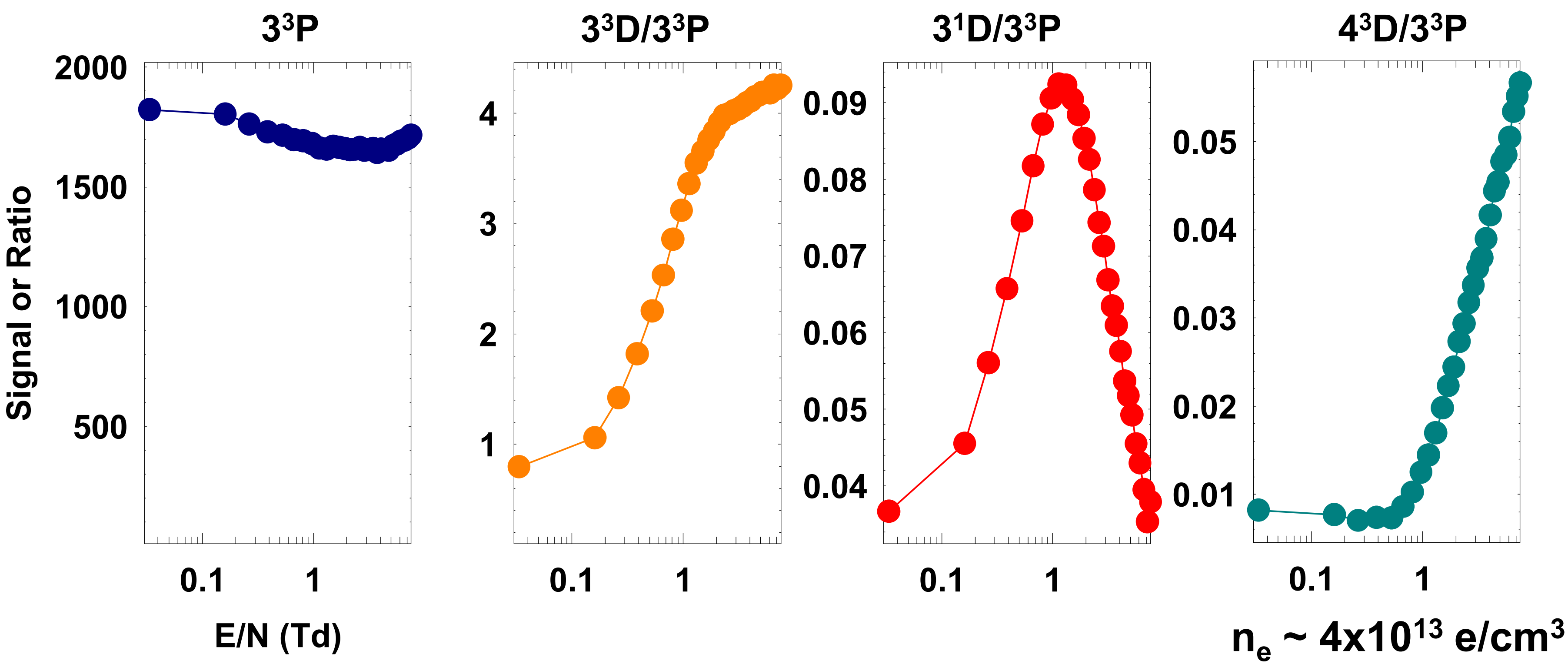
* Cross sections: Yu. Ralchenko, Atomic Data and Nuclear Data Tables **94**, 603 (2008)
†Characteristic energy: Bolsig and LXCat.

RATIO OF LCIF LINES ARE UTILIZED TO IDENTIFY SCALING TRENDS

- **E/N is tuned and LCIF is measured early during applied pulse.**
  - **Ratio of LCIF to laser excited 3^3P LIF is used to normalize signals.**
  - **LCIF acquired for ~ 10 ns, immediately after laser pulse.**

LCIF scaling with E/N



Several LCIF transitions are observable!

RATIO OF LCIF LINES ARE UTILIZED TO IDENTIFY SCALING TRENDS

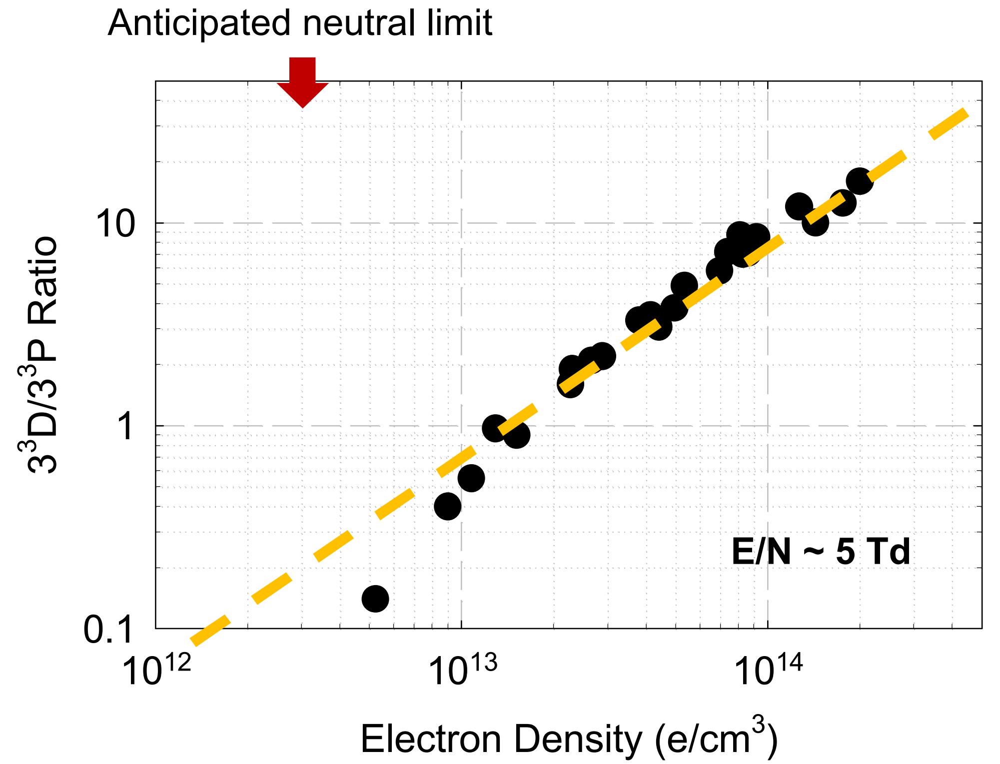
- **Benchmark scaling of density dependence of 3^3D LCIF.**
  - **Utilize the E/N invariance of 3^3D LCIF above ~ 2 Td**

Simplified Scaling

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

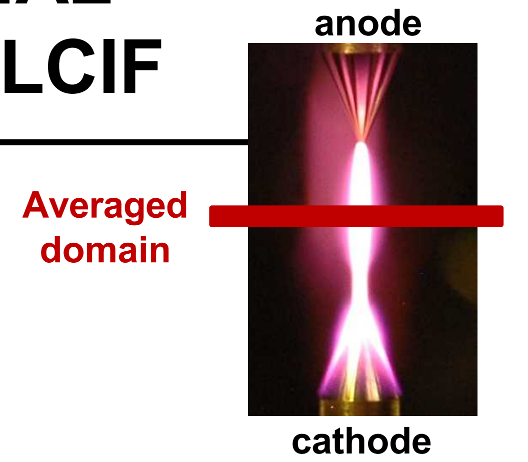
$$\frac{\Delta N_{3^3D}}{N_{3^3P}} \sim K_{ij}^{e} n_e$$

Observed Scaling of LCIF

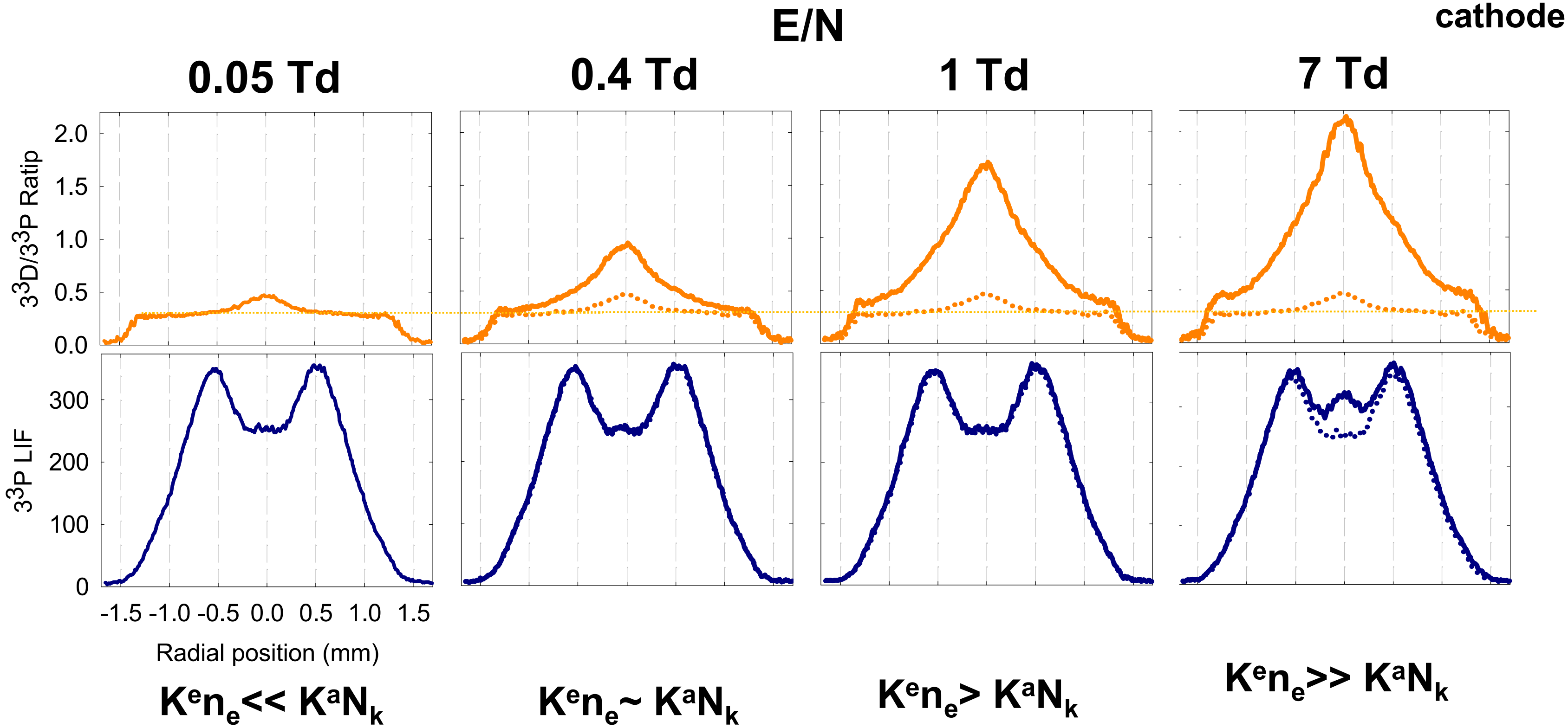


LCIF from 3^3D scales linearly with electron density

DEMONSTRATION OF SPATIAL RESOLUTION PROVIDED BY LCIF



- **Radial structure observed for various E/N.**
  - **Initial peak electron density of $2x10^{13}$ e/cm^3.**
  - **Measured ~ 2 μs in afterglow of first voltage pulse.**
  - **Measured in first 20 ns of applied heating pulse.**



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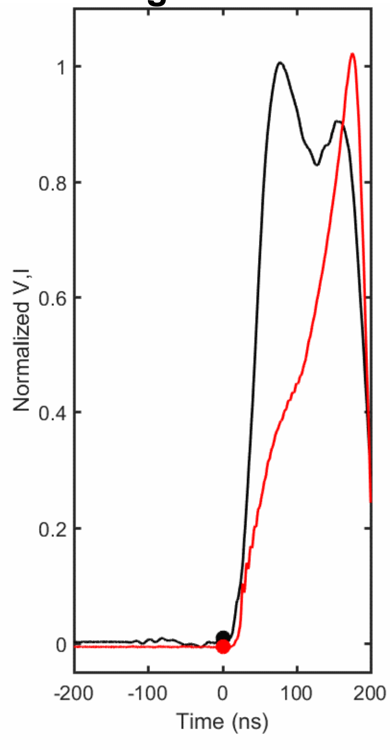
Observations consistent with initial assertions made

DEMONSTRATION OF SPATIALAND TEMPORAL RESOLUTION PROVIDED BY LCIF

- **Measured evolution of plasma formation during second pulse.**
  - **Initial peak electron density of $2x10^{13}$ e/cm^3 (2 μs afterglow)**

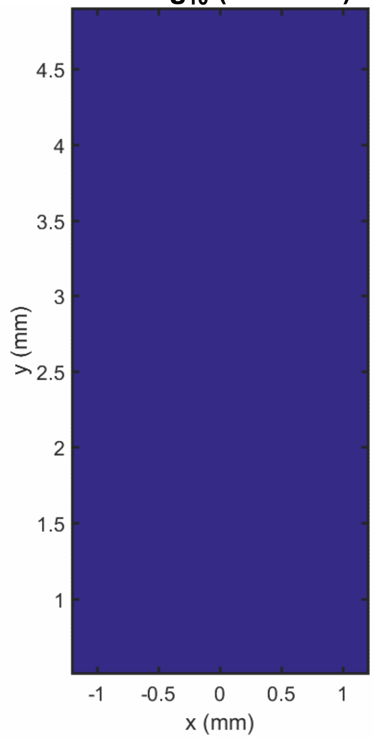
Waveforms

Voltage Current



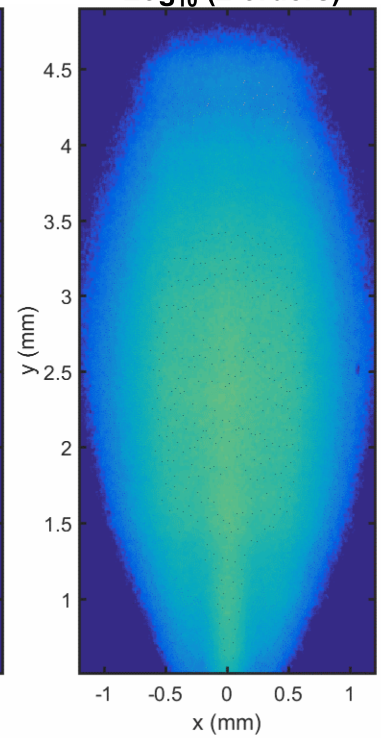
Emission

Log_{10} (2 orders)



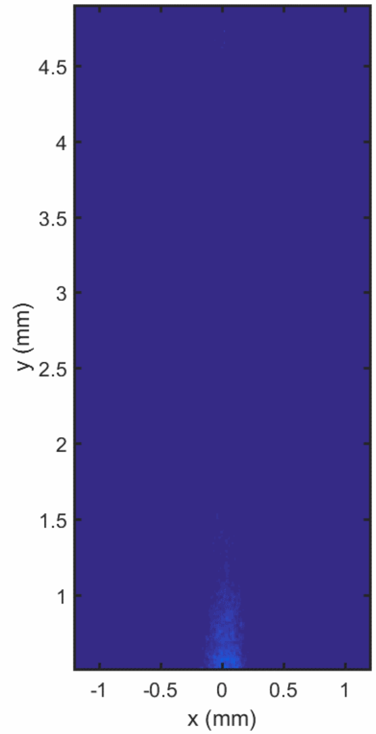
Metastables

Log_{10} (2 orders)

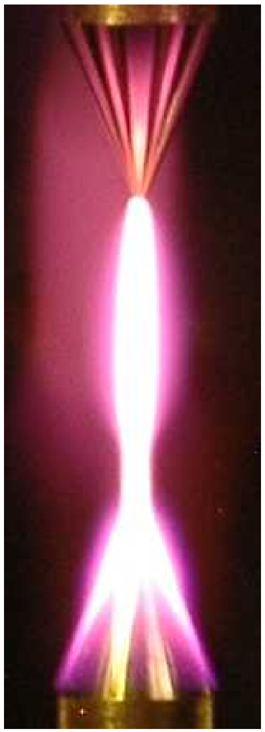


Electrons

Linear ($2x10^{14}$ e/cm^3 peak)



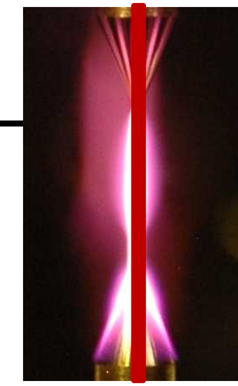
anode



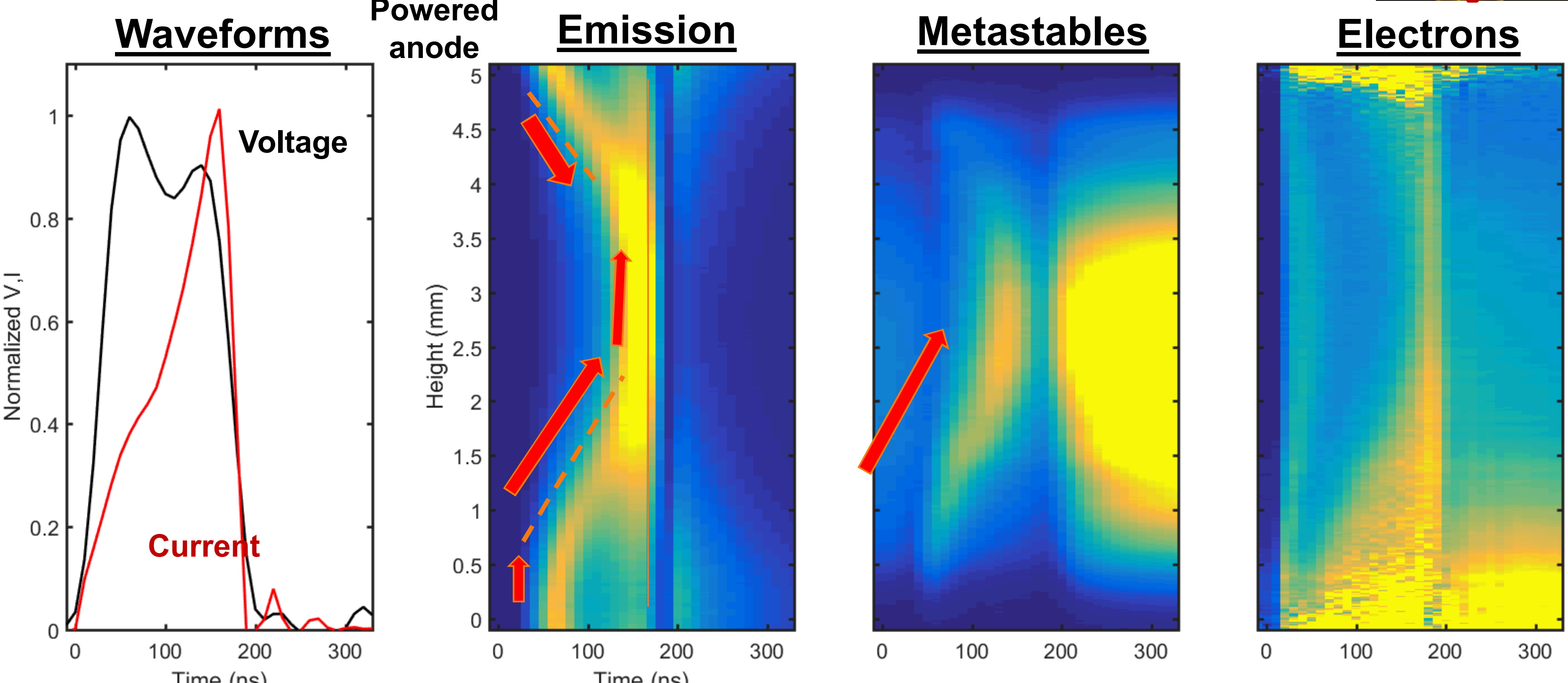
cathode

LCIF provides two-dimensional plasma structure

UTLIZATION OF LCIF TO UNDERSTAND DISCHARGE EVOLUTION



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



LCIF captures spatial and temporal evolution of plasma formation.

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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!

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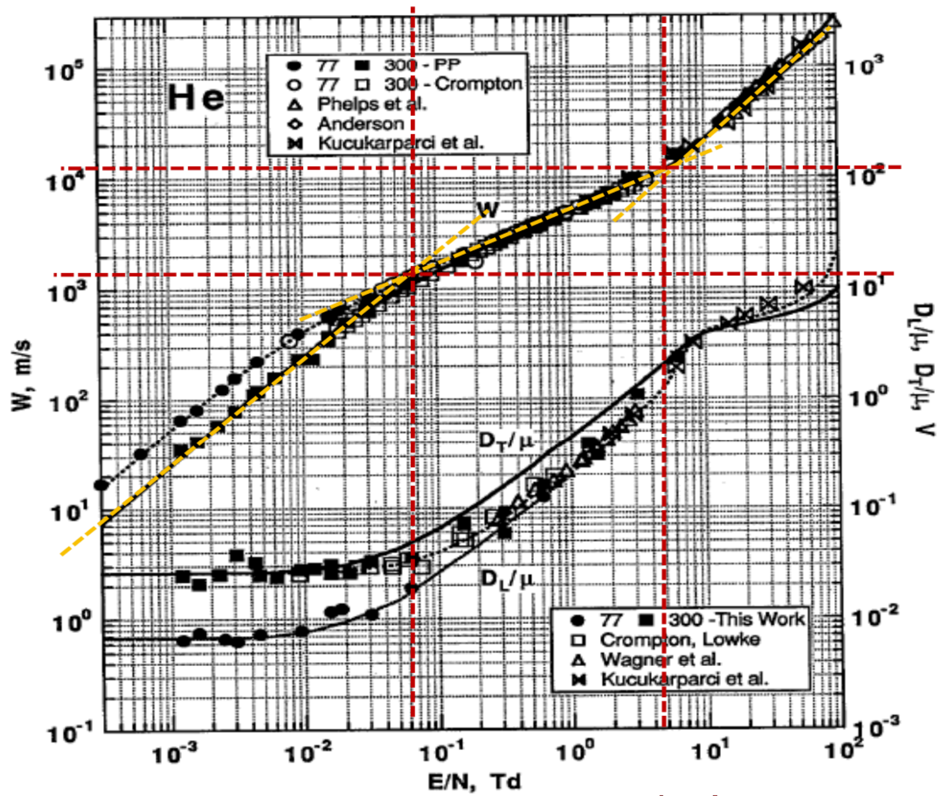
Contracts DE-SC0001939 and DE-AC04-94SL85000

AUXILARY SLIDES

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