



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

**15th International Symposium on
Laser-Aided Plasma Diagnostics (LAPD)
Jeju, Korea, October 9'th - 13'th 2011**

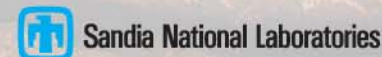
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Laser-collision induced fluorescence provides measure of electron density and "temperature"

- **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

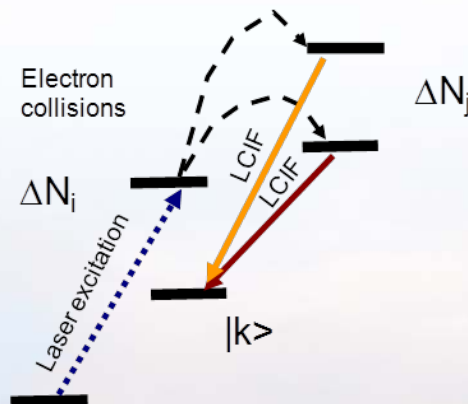


Part I: LCIF concepts and key trends

- **Overview**

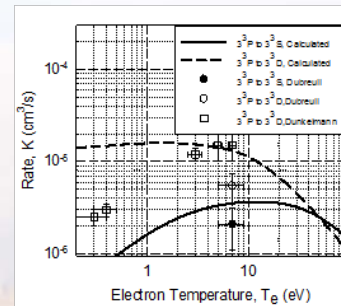
- LCIF concepts
- Collisional-radiative model for helium
- Key scaling trends

LCIF Concept

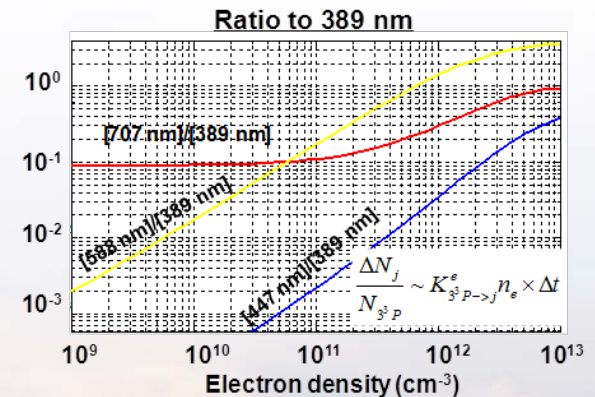


CR Model

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

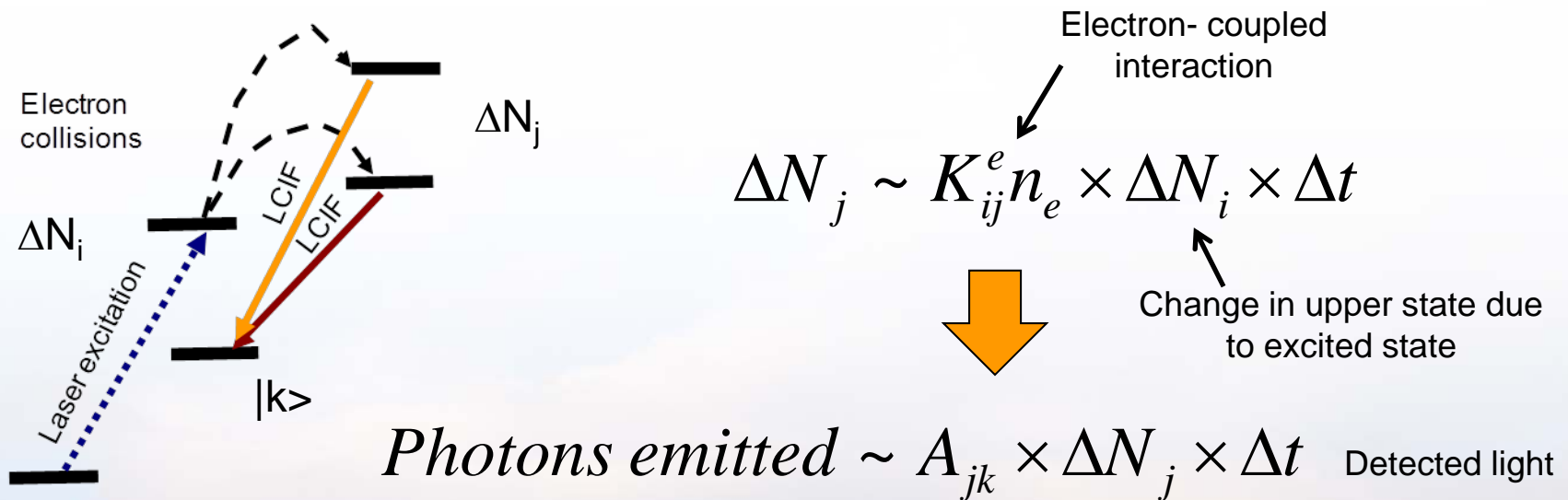


Key scaling trends



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



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

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

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

Electron-temperature dependent rates

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

Distribution function used for describing electron velocities

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

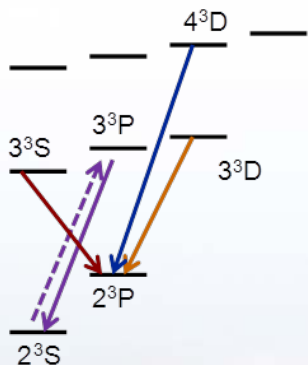
Approach is applicable to various atomic and molecular systems of interest



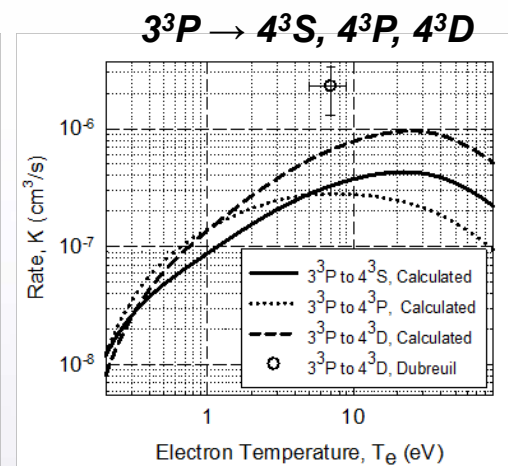
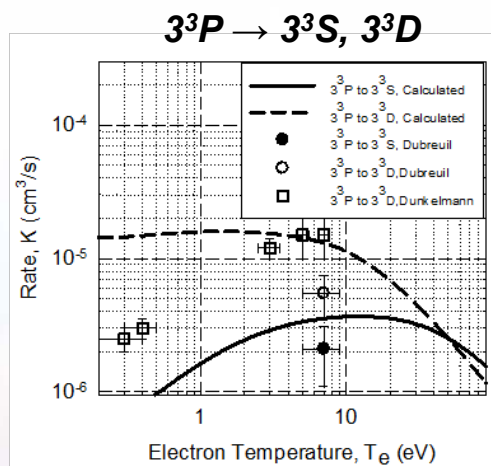
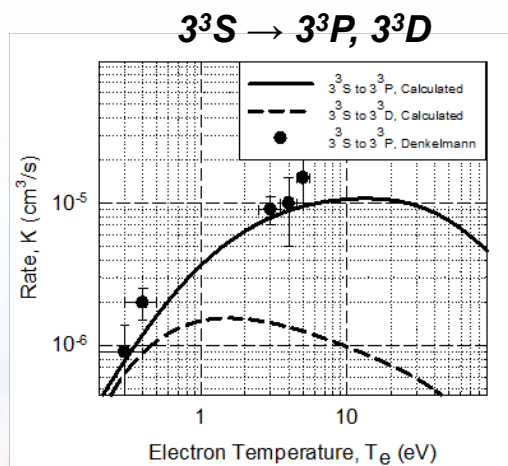
Helium atom serves as target species for LCIF measurements

- Employ Helium to start with - considering argon
 - "Simple system" with "better known" rates
- Utilize functionalized form of cross-sections compiled by Ralchenko¹
 - Integrate to get rates, compare to measured rates^{2,3}

Key transitions



Computed and measured excitation rates in Helium



- Yu. Ralchenko, R. K. Janev, T. Kato, D. V. Fursa, I. Bray, F. J. De Heer, Atomic Data and Nuclear Data Tables **94**, 603 (2008)
- R. Denkelmann, S. Maurmann, T. Lokajczyk, P. Drepper, and H. -J. Kunze, J. Phys. B: At. Mol. Opt. Phys. **32**, 4635 (1999).
R. Denkelmann, S. Freund and S. Maurmann, Contrib. Plasma Phys. **40**, 91 (2000).
- B. Dubreuil and P. Prigent, J. Phys. B: At. Mol. Opt. Phys. **18**, 4597 (1985).



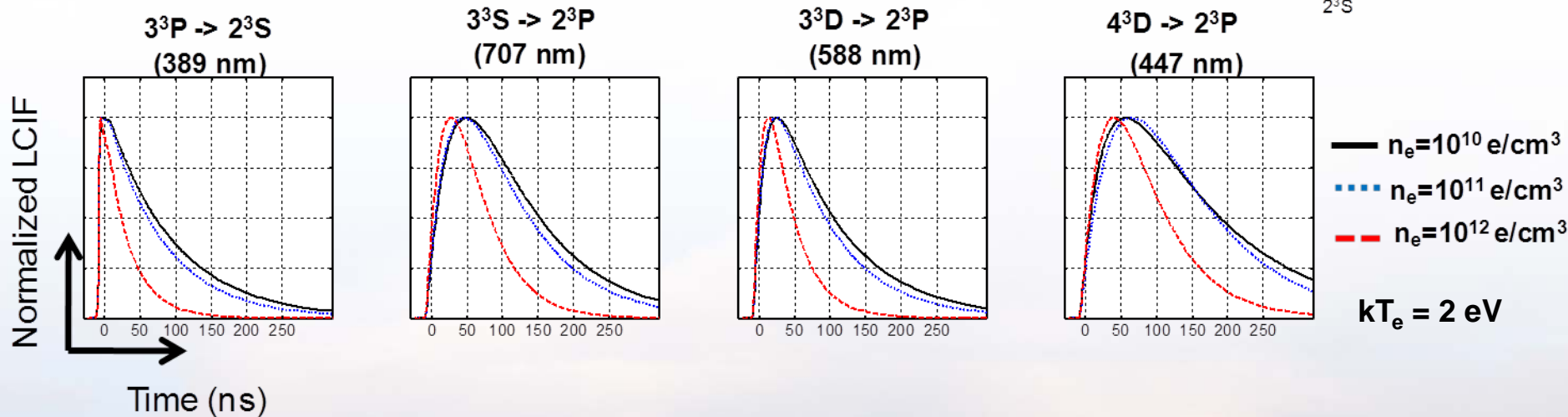
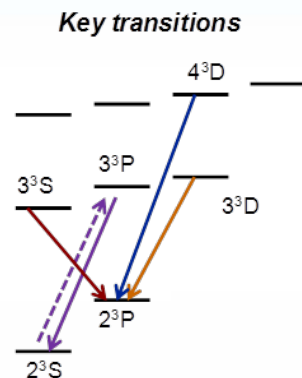
Accuracy of n_e , T_e depend on knowledge of $K_{ij}(kT_e)$

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/cm³
 - Below $n_e \sim 10^{11}$ absolute intensities are needed

$$K_{ij}^e n_e \sim A_{ij} \quad \Rightarrow \quad 10^{-5} \times 10^{11} \sim 10^7$$

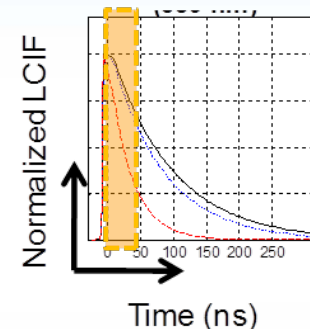
Computed temporal evolution



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

Window of integration



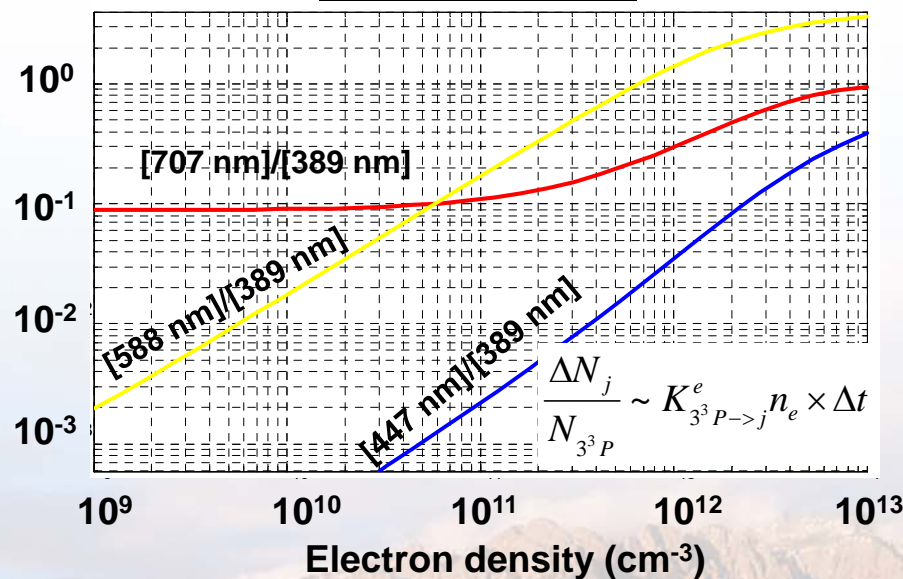
Examine ratios of time integrated LCIF

- Eliminates need for absolute calibrations
- Still need relative efficiencies of imaging system

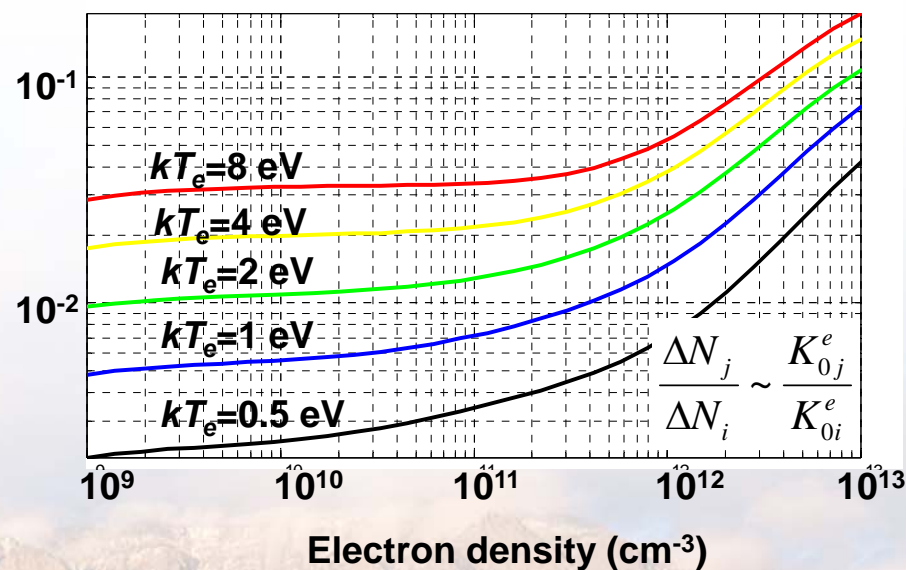
Capitalize on " kT_e independent" coupling of 3^3P to 3^3D

- Ratio of 588 nm to 389 nm yields n_e
- Density + Ratio of 447 nm to 588 nm yields kT_e

Ratio to 389 nm



Ratio [447 nm]/[588 nm]

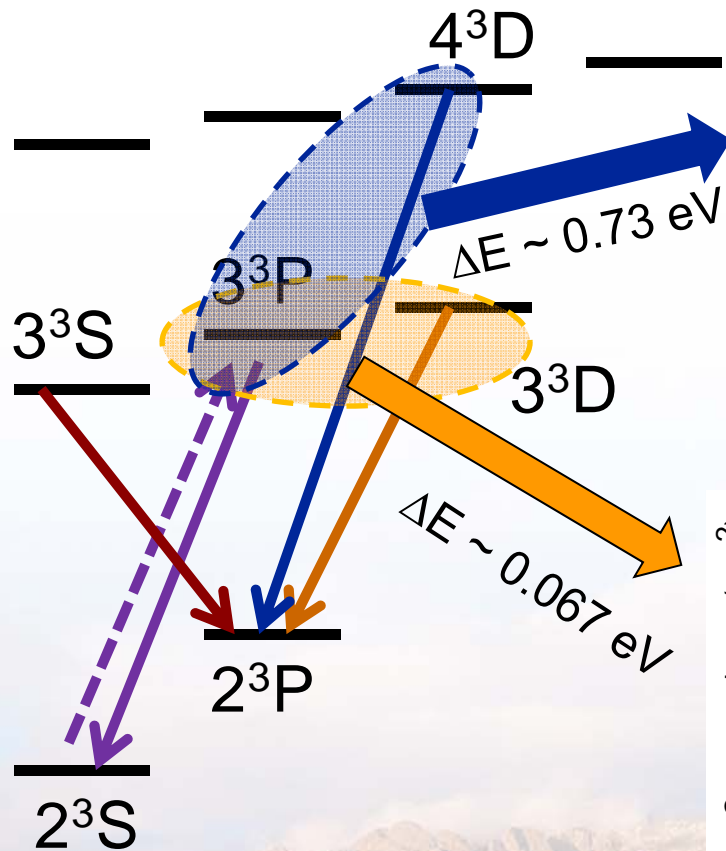


Only need to make three measurements to obtain n_e , kT_e

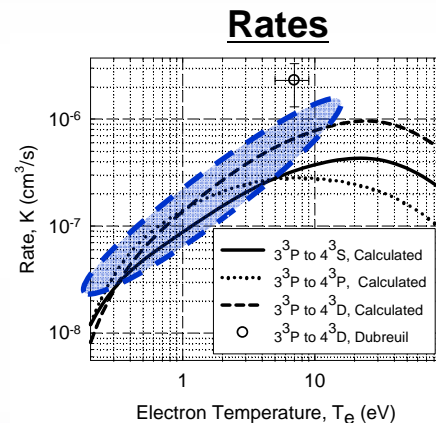
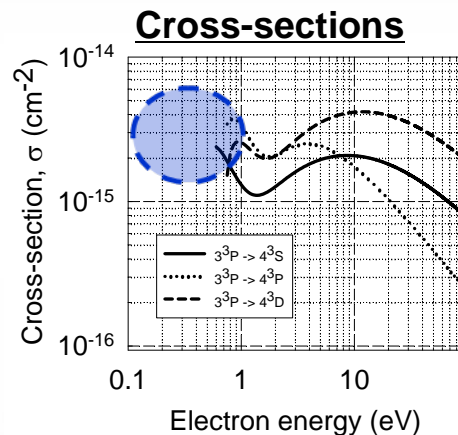


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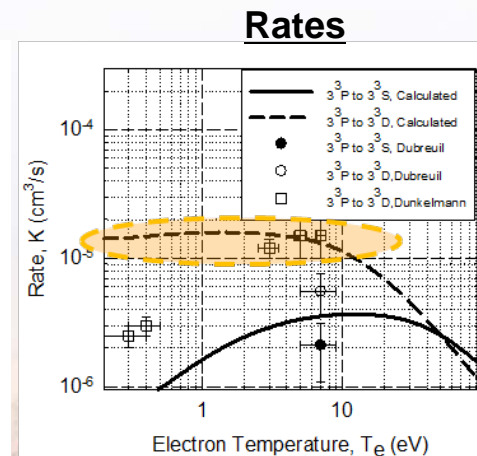
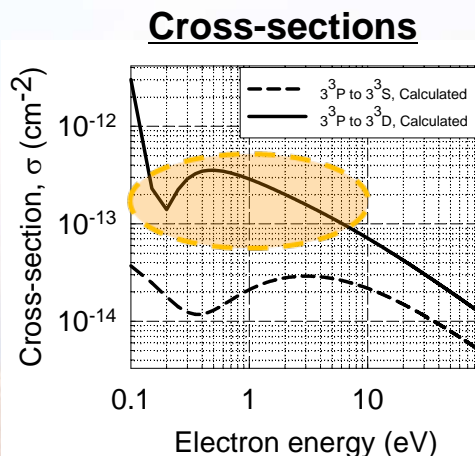
Small energy gap leads to " kT_e independent" coupling of 3^3P to 3^3D



$3^3P \rightarrow 4^3D$



$3^3P \rightarrow 3^3D$



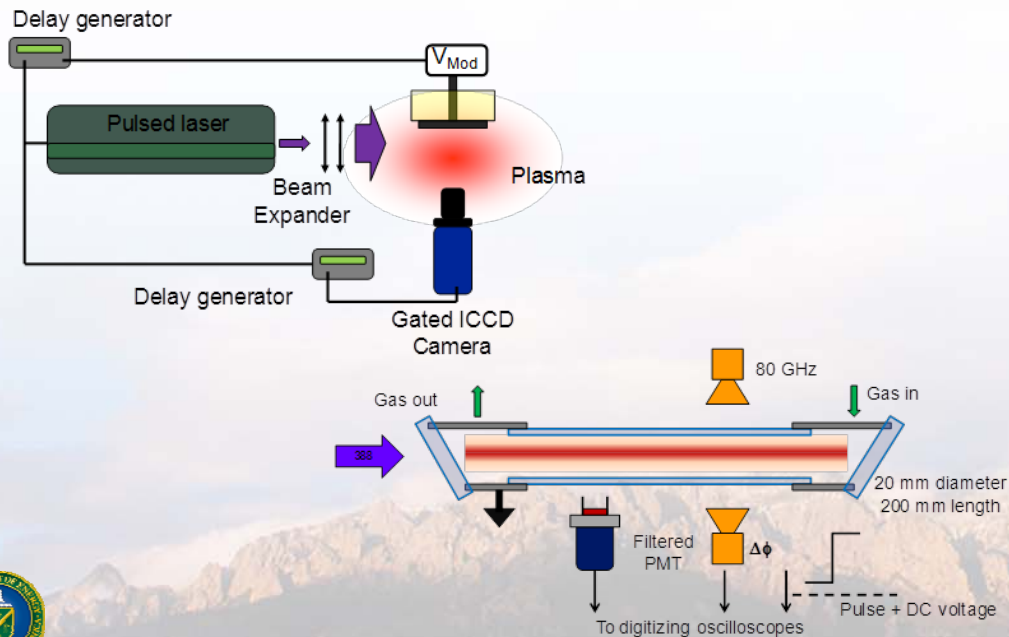
Considerable fraction of the electrons are capable of driving the interaction



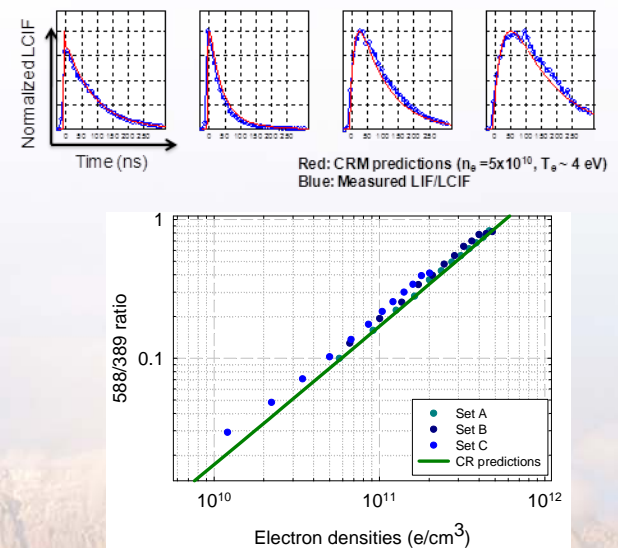
Part II: LCIF implementation and benchmark

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

Experimental setup



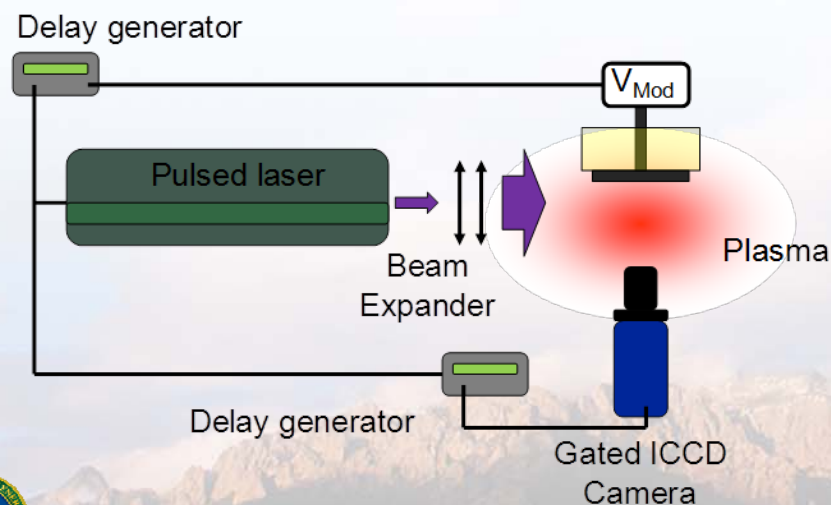
Benchmark LCIF



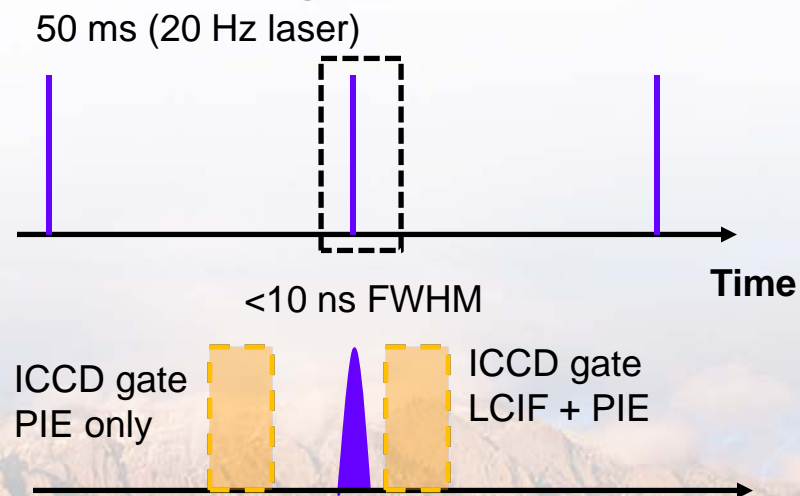
Experimental implementation of the LCIF is realized

- **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 (3×2) measurements to obtain n_e , kT_e

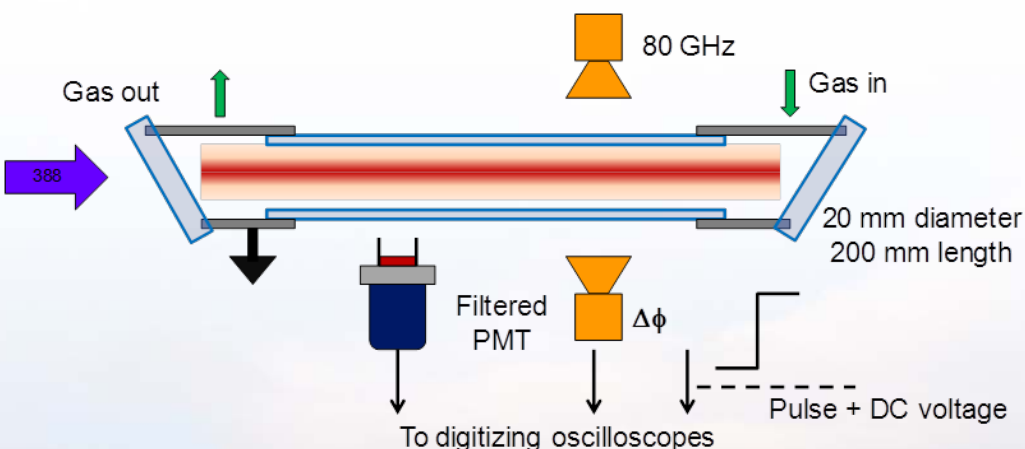


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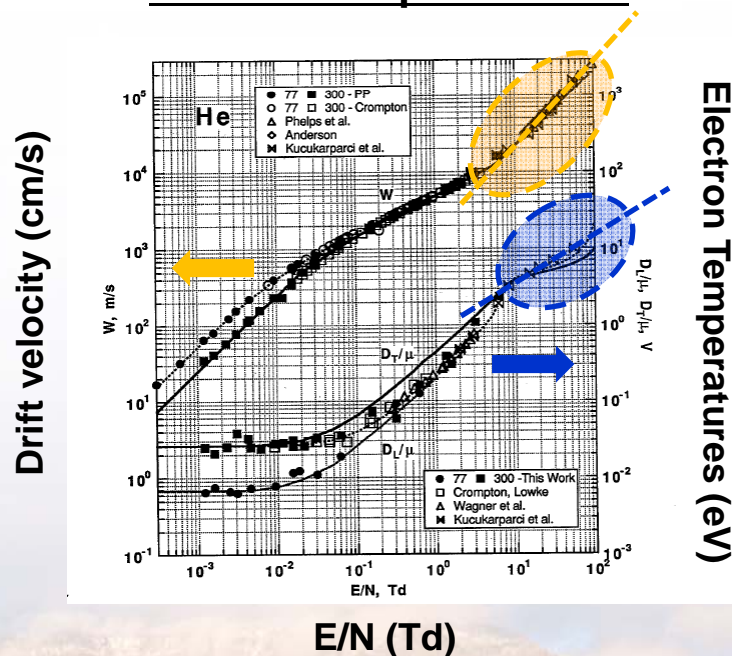
Pulsed positive column is utilized to benchmark LCIF technique

- 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



Helium drift parameters



Positive column is a good vehicle to benchmark LCIF technique

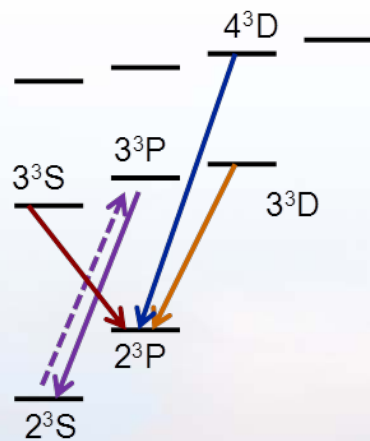


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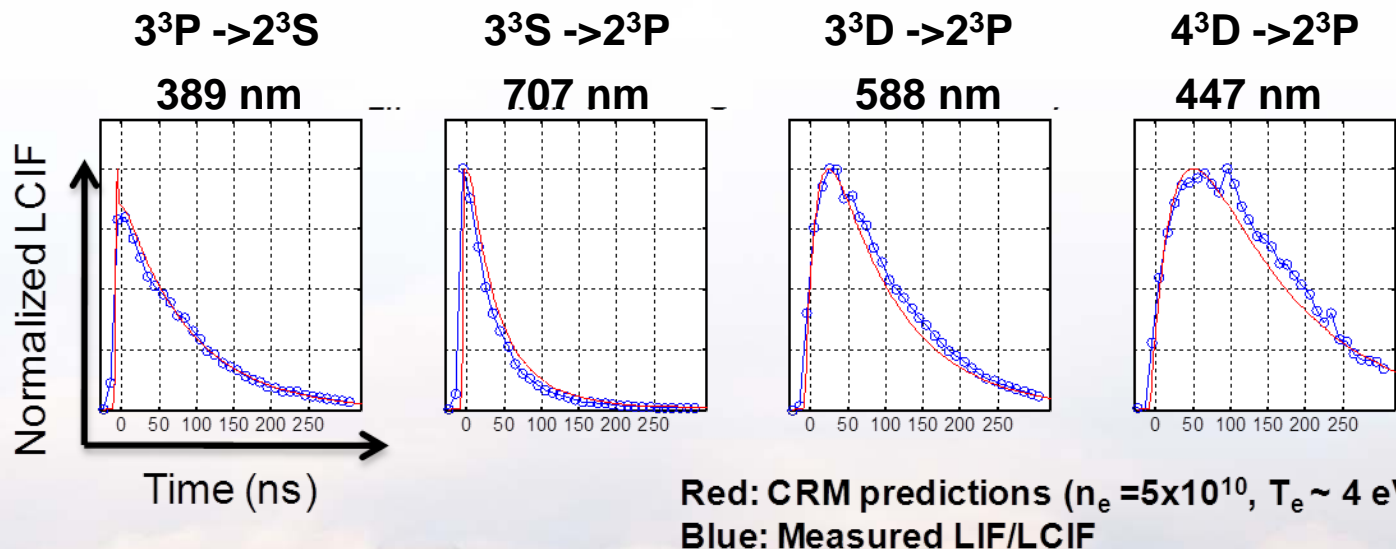
First steps: Verify time resolved LCIF to test CRM

- 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



Representative results



Good first demonstration of LCIF technique

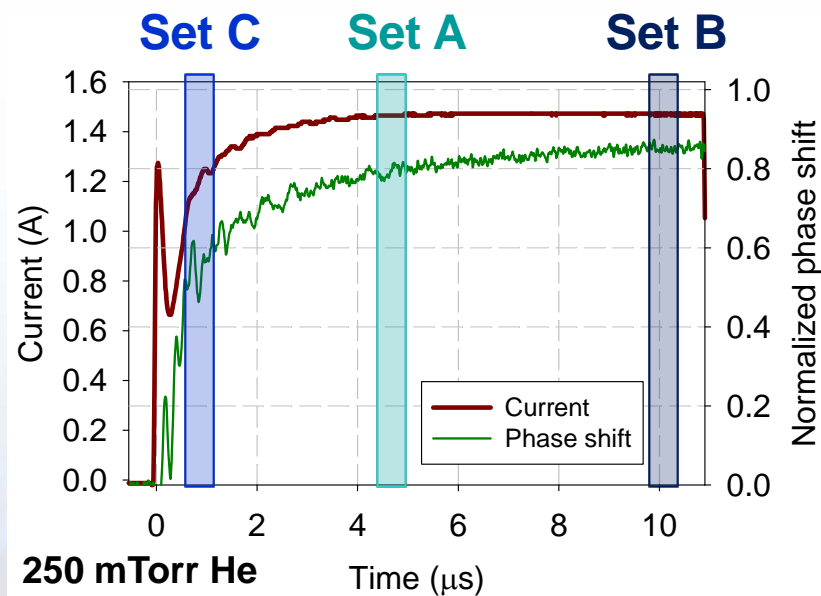


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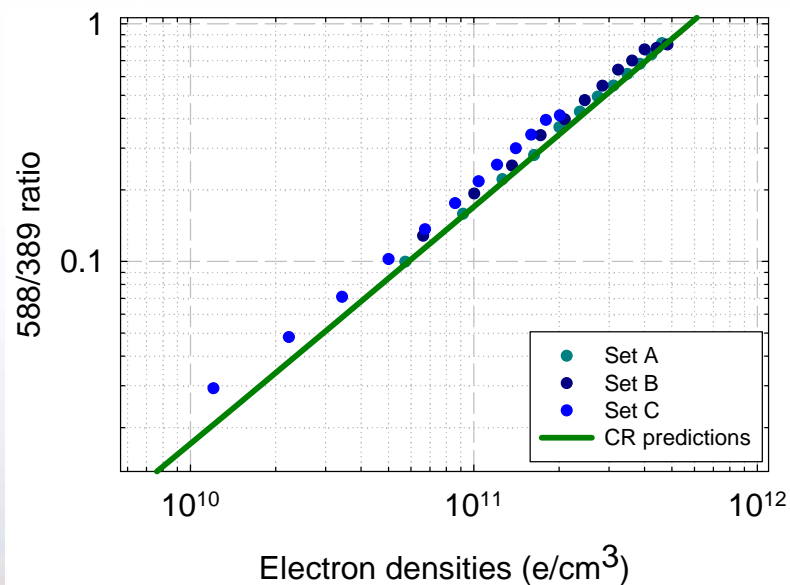
[588]/[389] ratio exhibits linearity over nearly two orders of magnitude

- 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

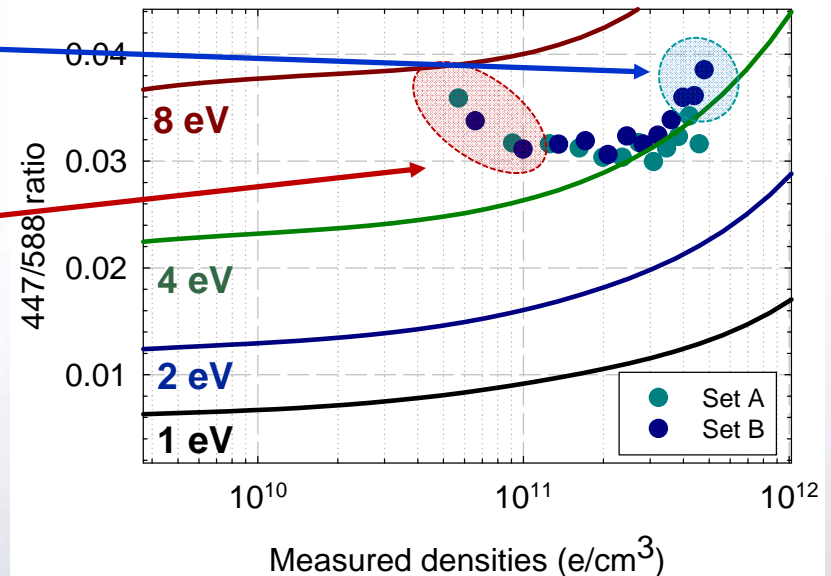
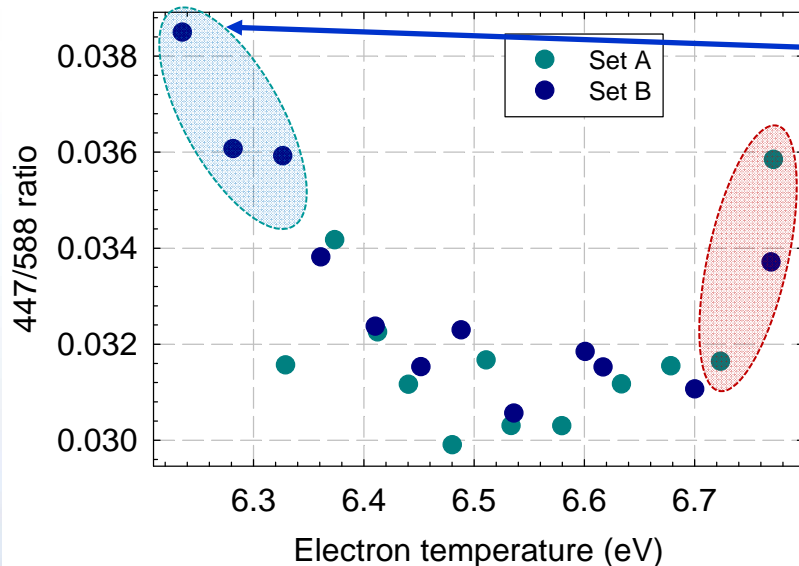
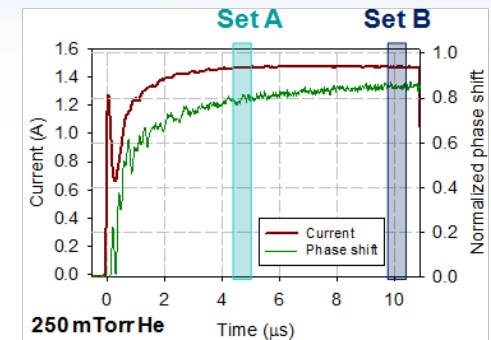


*Density measurements obtained at different times
essentially overlay each other*



[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



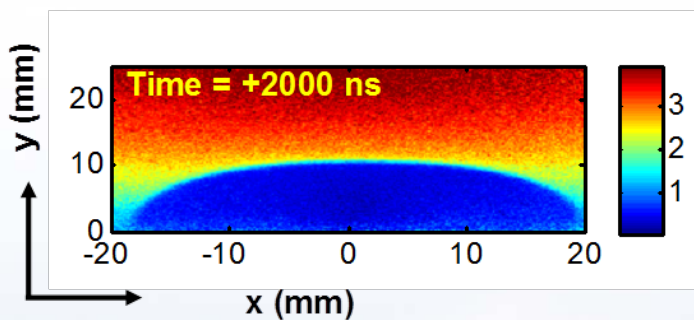
Uncertainties in rates, EEDF and/or interpolation of T_e from drift parameters should impact absolute values



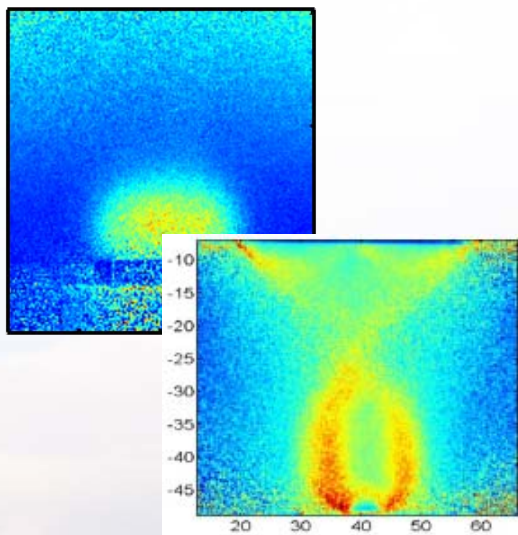
Part III: Applications of LCIF

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

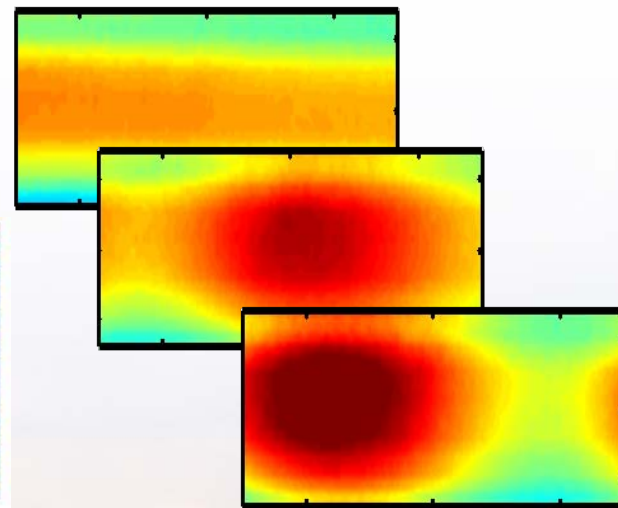
Ion sheath



Double layers



Striated positive column



Emphasize structure and evolution of the plasma being studied

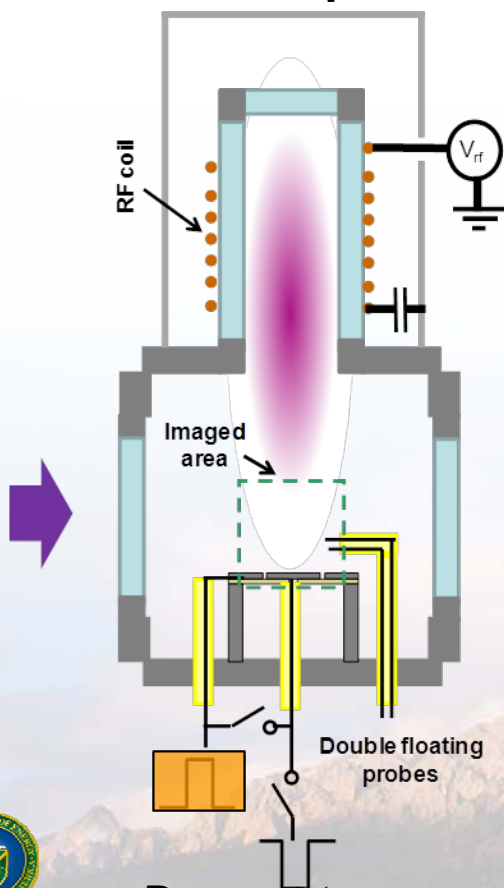


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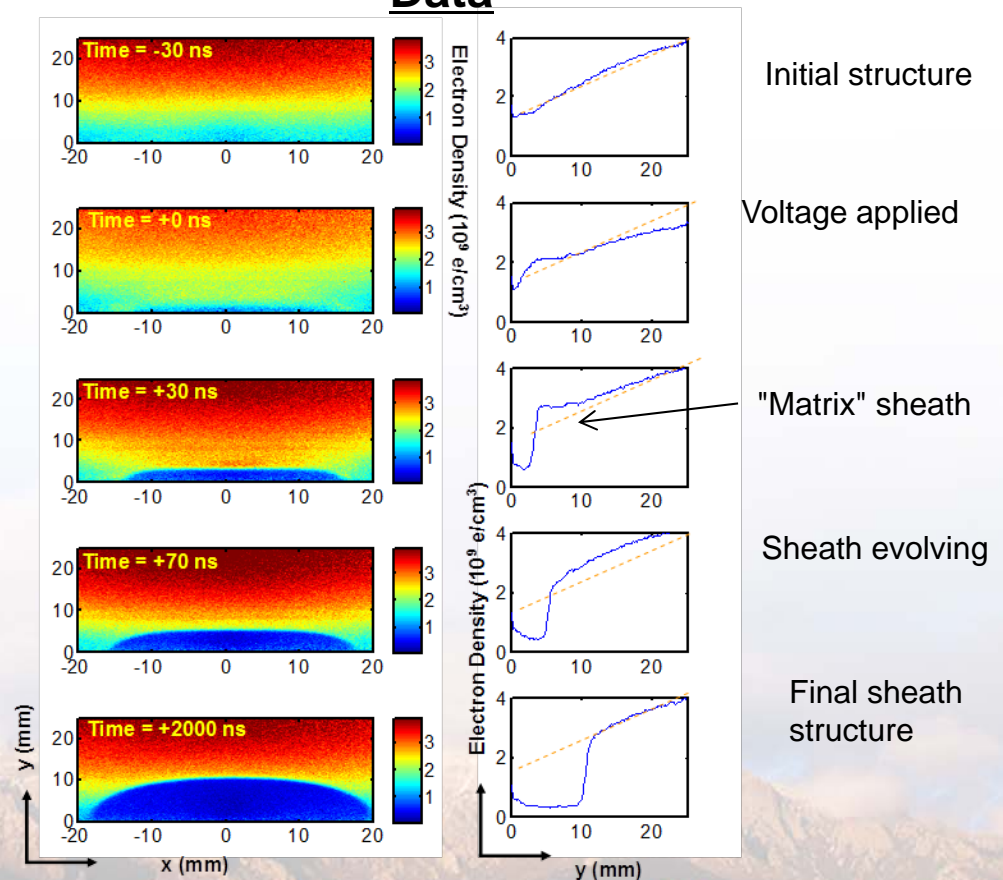
Demonstration of LCIF technique: 2D-sheath formation

- 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

Setup



Data



Decent temporal and spatial resolution demonstrated



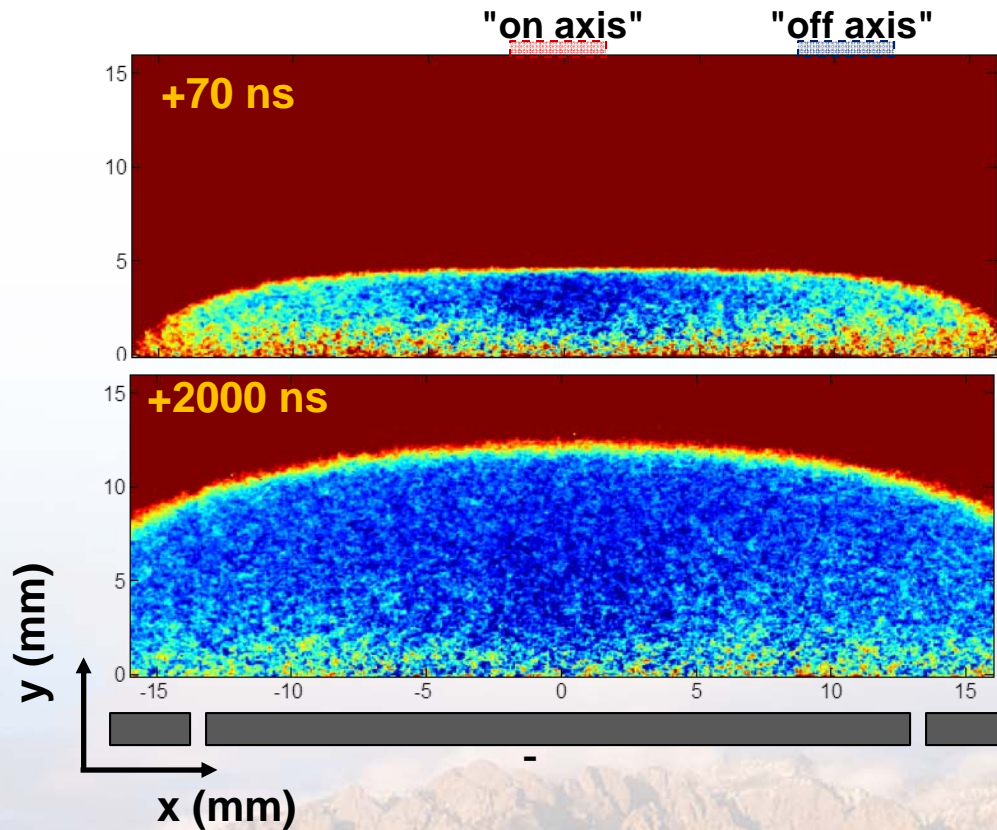
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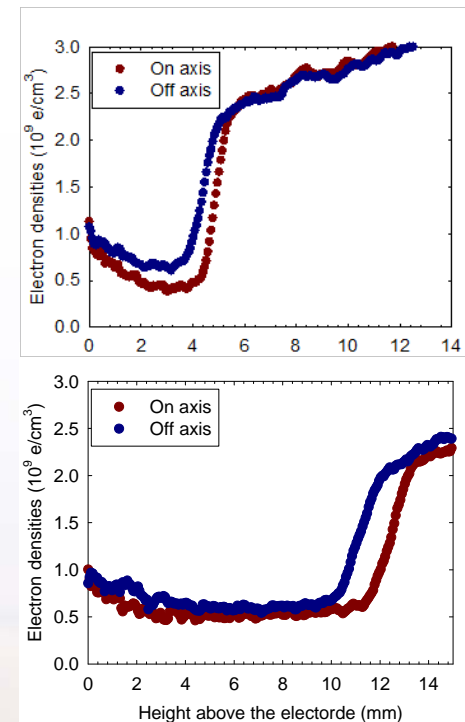
Interesting structure observed in the sheath

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

2D Images



Profiles

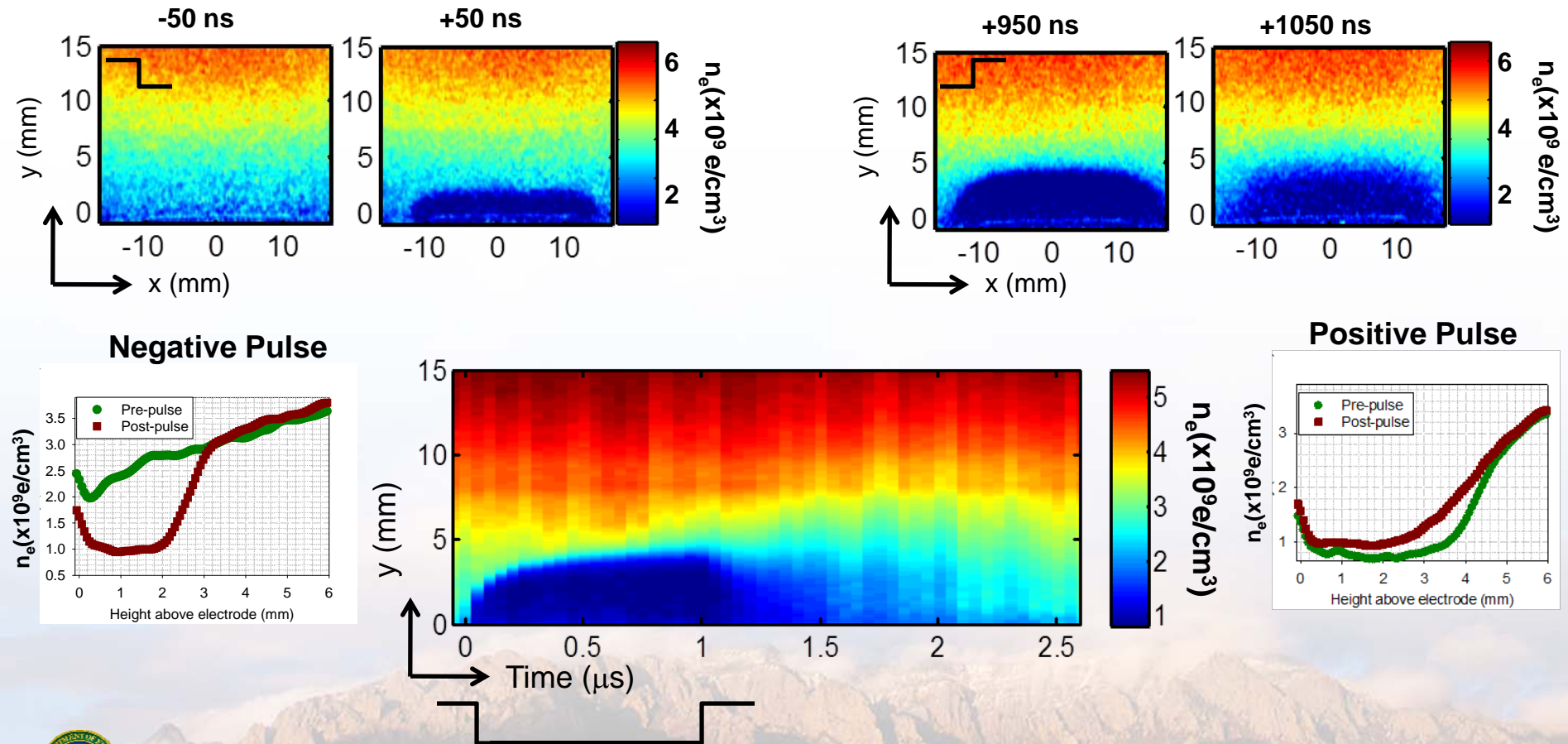
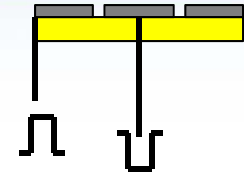


Signal deep in the sheath caused by electrons emitted from the electrode



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

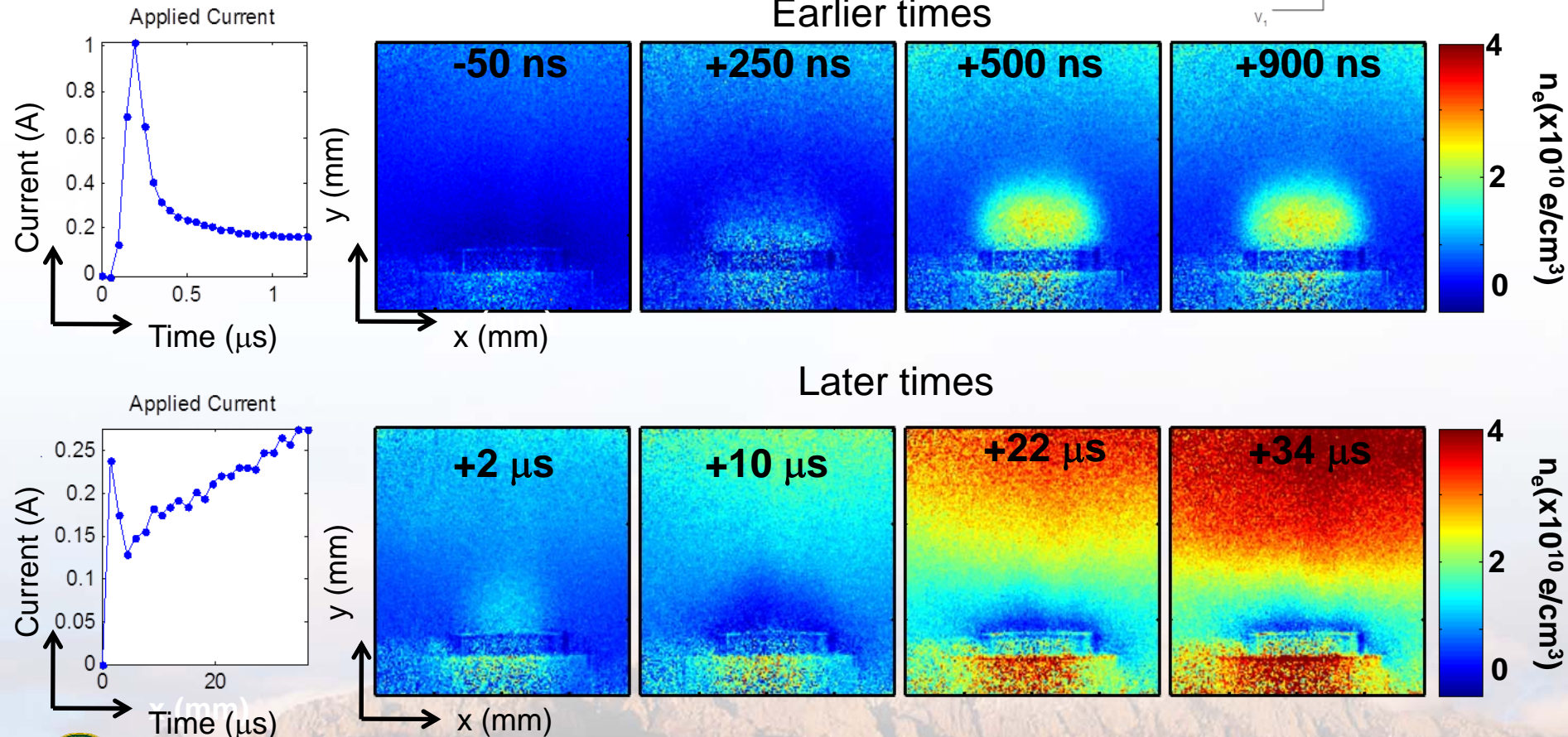
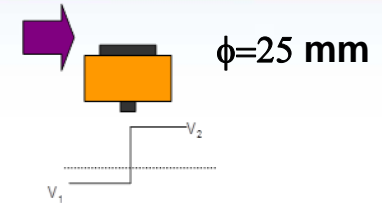


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Transient anodic double layer observed after pulsed excitation

- Closer analysis of initial plasma distribution

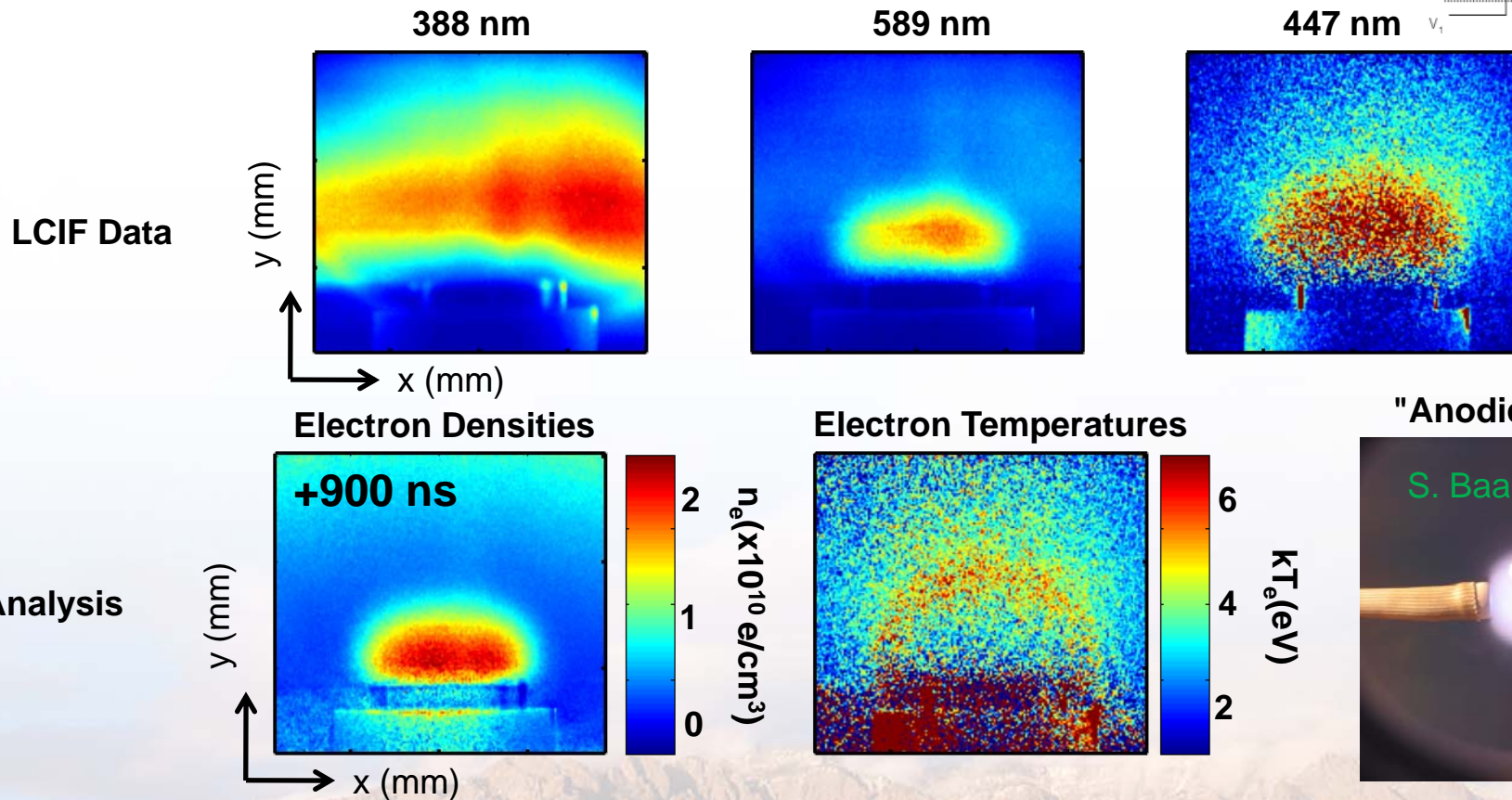
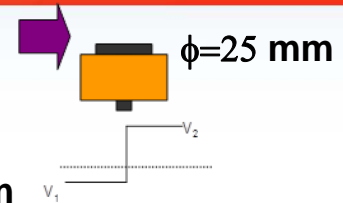
- Use smaller (25 mm) diameter electrode, 100 mTorr afterglow



Structure undergoes inversion after stagnation

Higher energy electrons observed around edge of anode plasma

- Temperature measurements made for +900 ns case
 - Challenging measurement because of low level signals



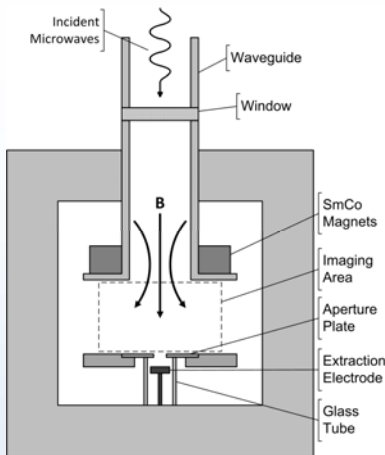
*Electrons energized by localized electric fields
supporting double layer*



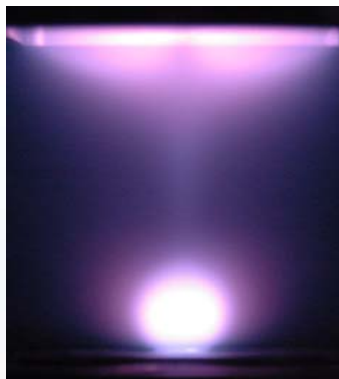
Double layer more pronounced in ECR based plasma cathodes

- **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

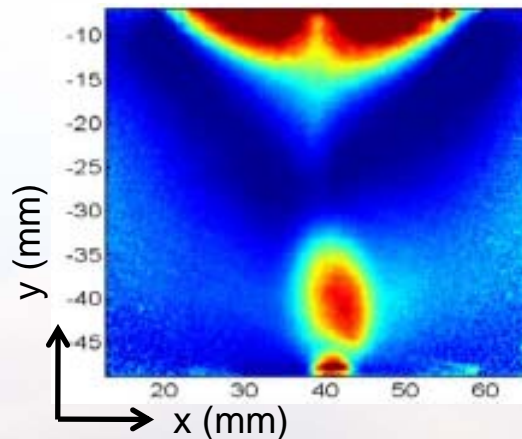
Setup



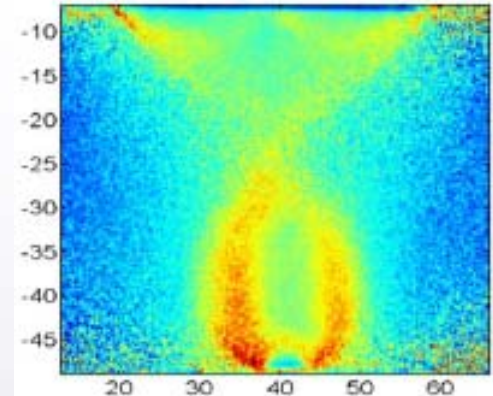
"Full color" picture



Density



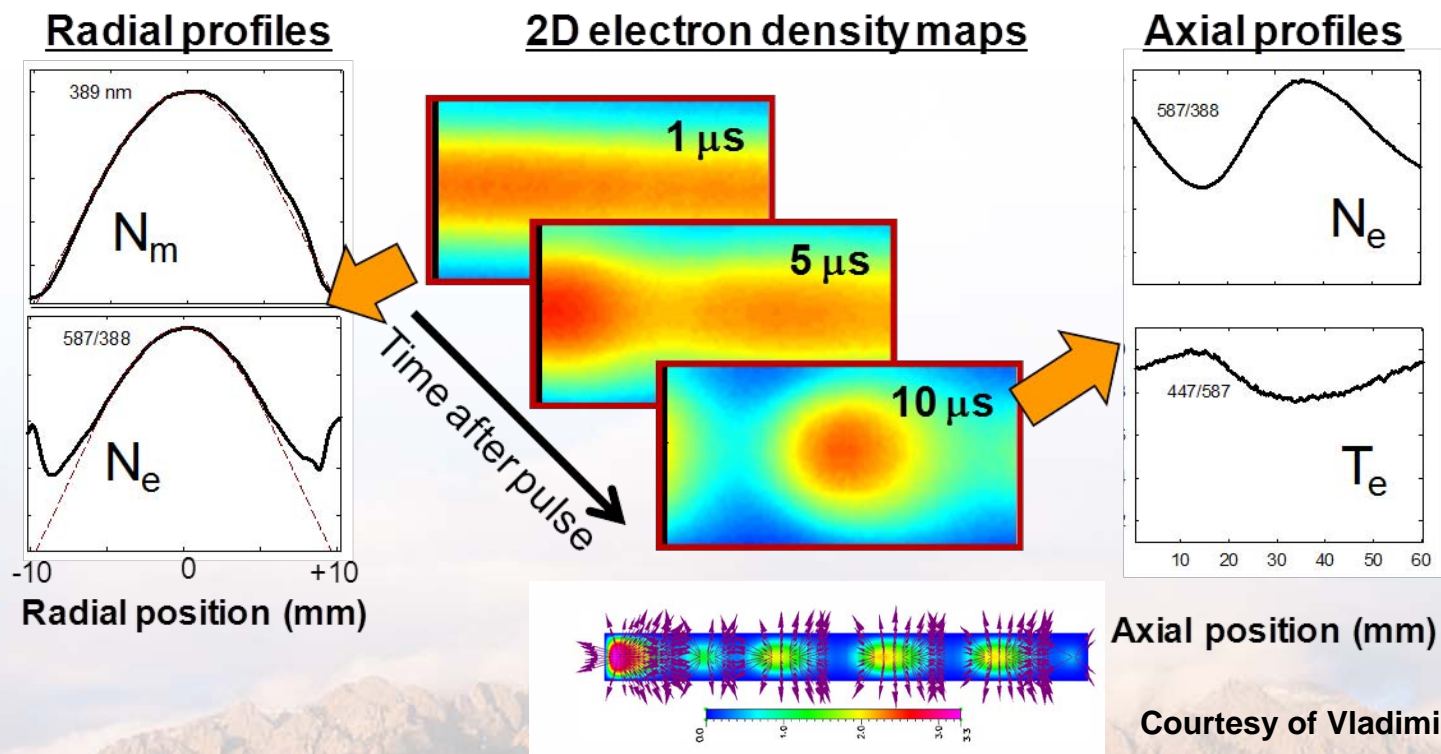
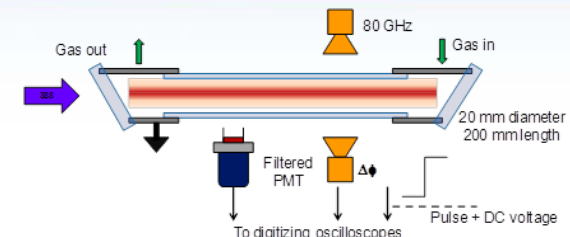
Temperature



***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



Courtesy of Vladimir Kolobov

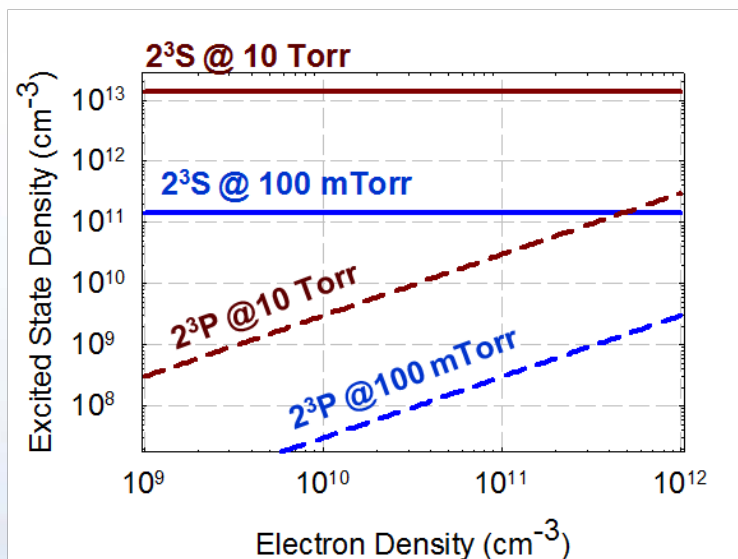
**Benchmark 2D simulations with measurements
made by LCIF**



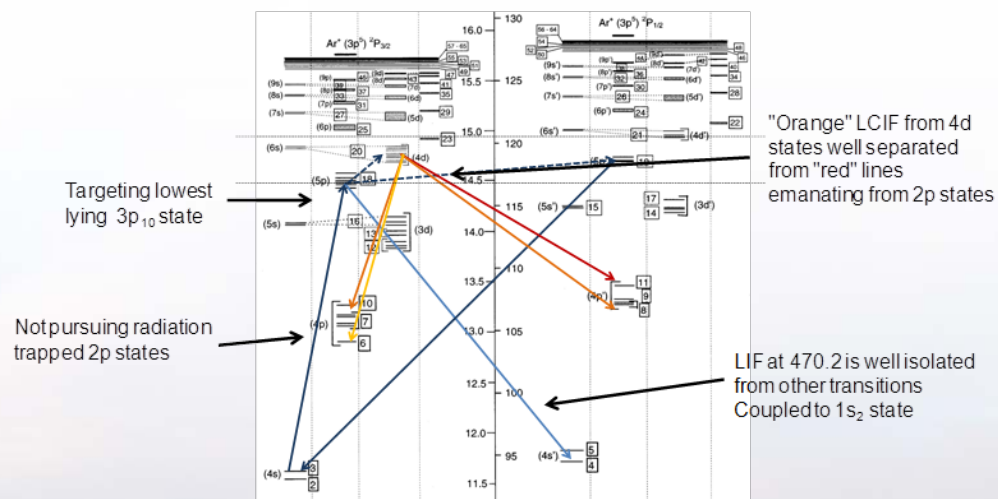
Part IV: Future pursuits with LCIF

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

Extension of helium



Argon LCIF



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

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

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

(Assuming $L=1$ cm and $N_A \sim 10^{13}$ absorbers/cm³)

States connected to 2^3S can become trapped do to strong coupling and higher populations



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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

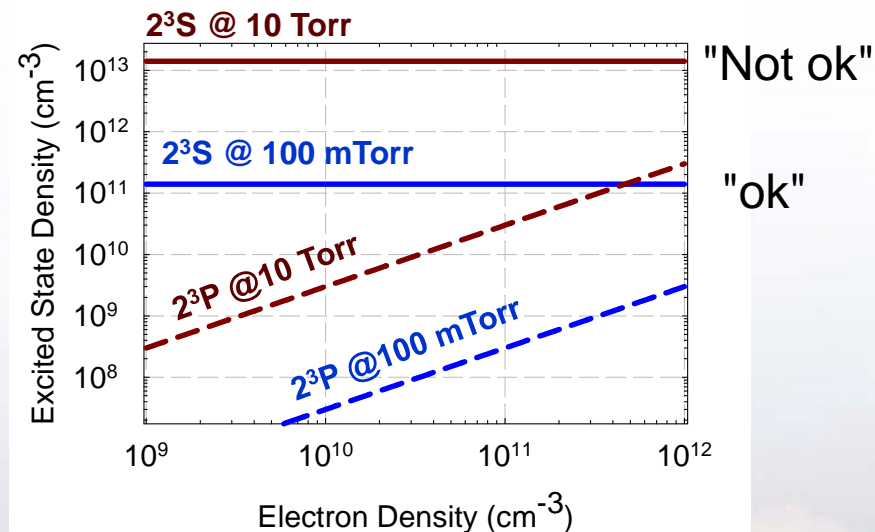
2^3S State:

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

2^3P State:

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

Trends



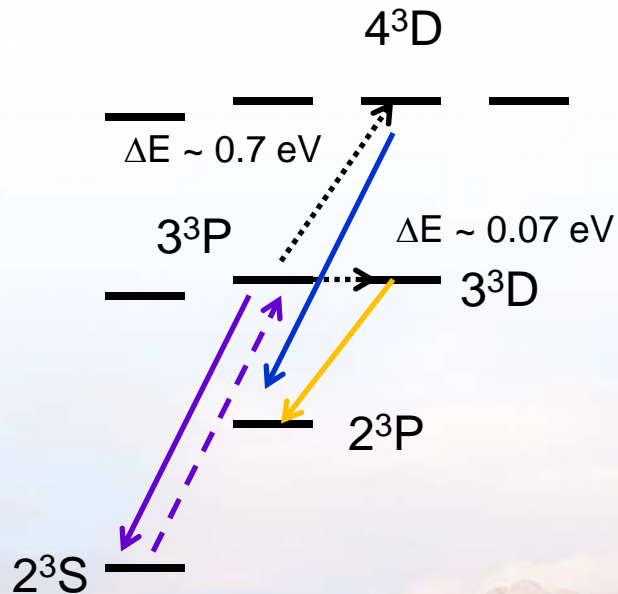
Low 2^3P state densities should be high enough to pump but low enough not to trap



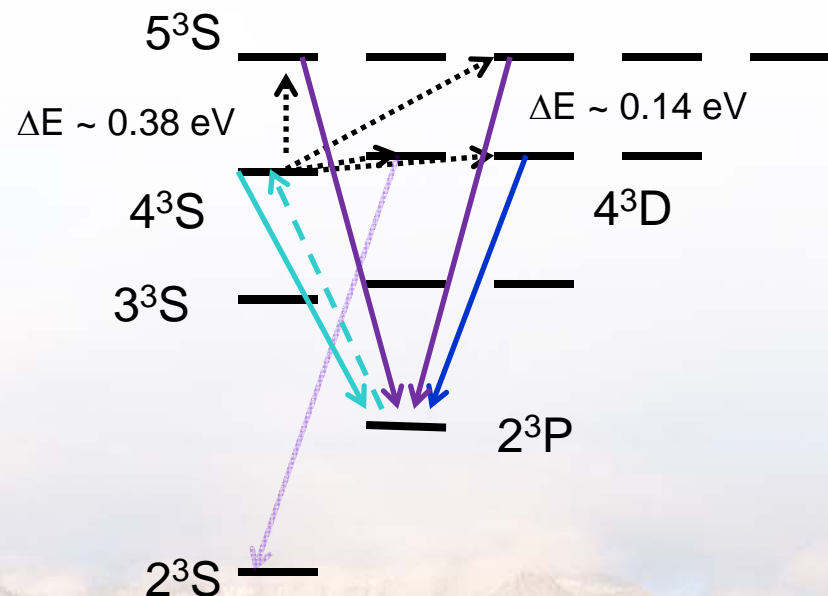
Spectroscopic pathway proposed for pumping from 2^3P

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

Previous approach



Proposed approach



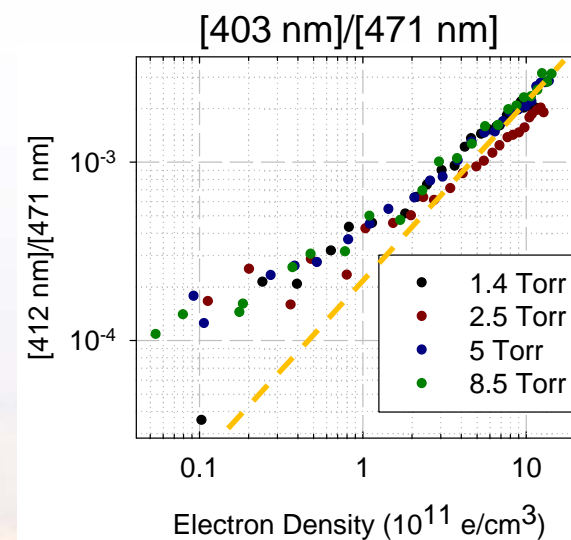
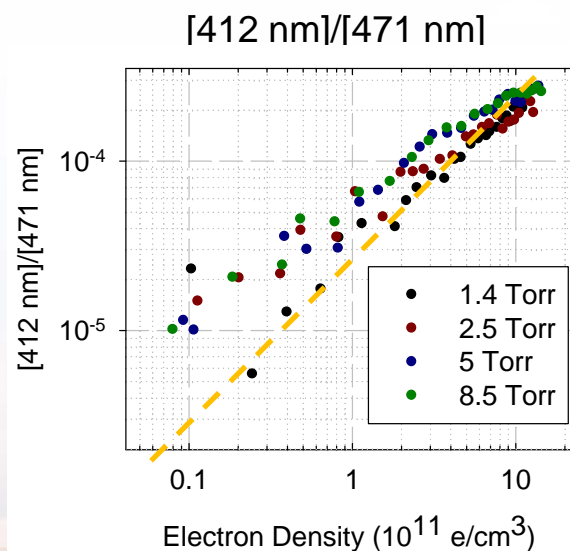
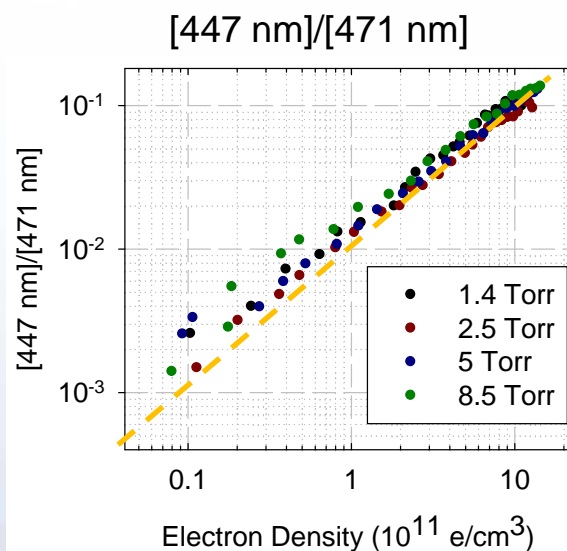
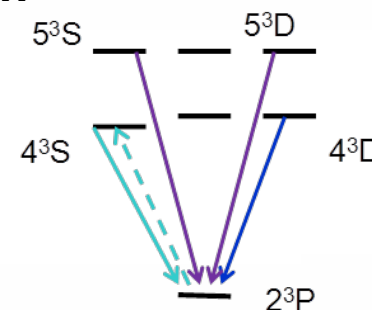
Pumping to the 3^3S or the 3^3D states are not "off the table" and may be perused



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



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

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

Helium

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

Argon

Transition	f_{nm}	τ
$2p_{10} \rightarrow 1s_5$	0.17	$>>1$
$3p_{10} \rightarrow 1s_5$	9×10^{-4}	0.08

(Assuming $L=1$ cm and $N_A \sim 10^{13}$ absorbers/cm³)

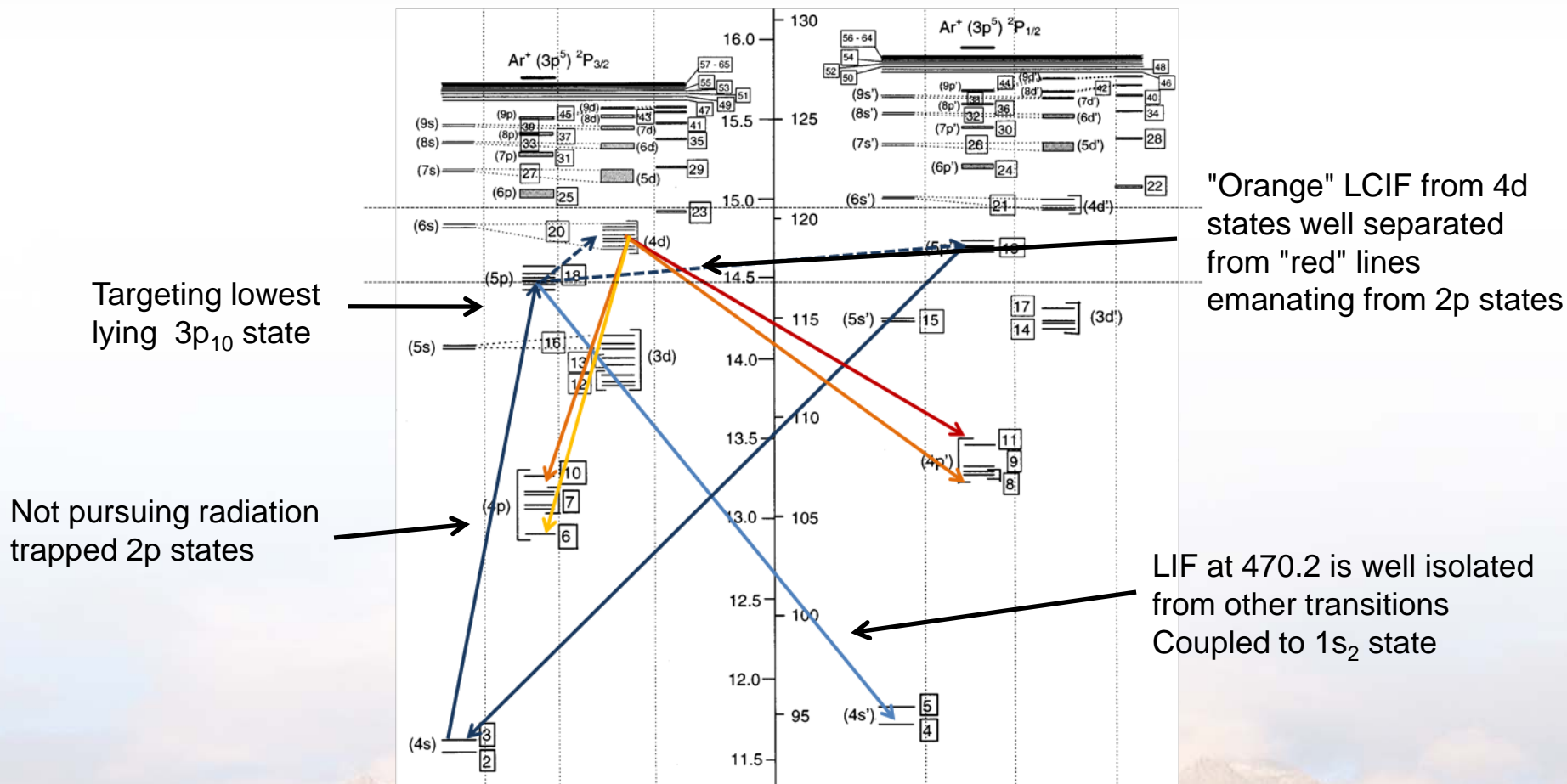
Low oscillator strengths and possibly lower densities make argon attractive



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Complexity of argon makes extension of LCIF "challenging"

- Argon offers more spectroscopic pathways to pursue



Taken from Bogearts et. al, J. Appl. Phys. **84**, 121, 1998

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

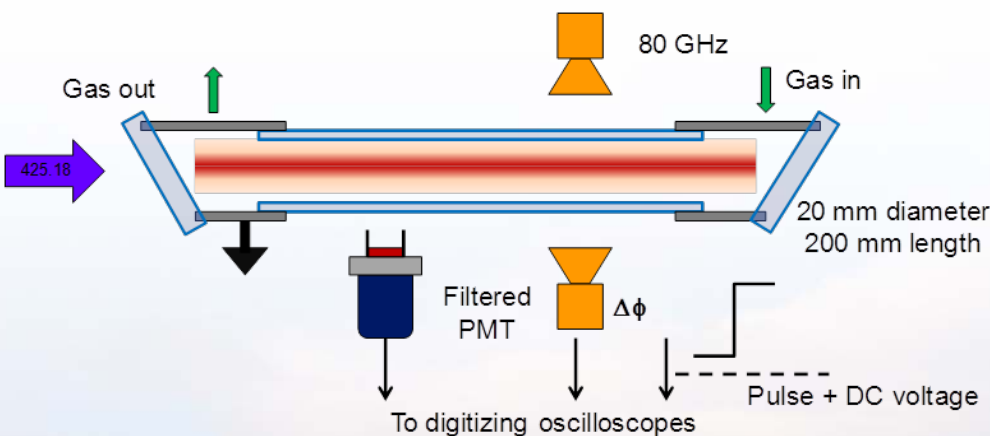


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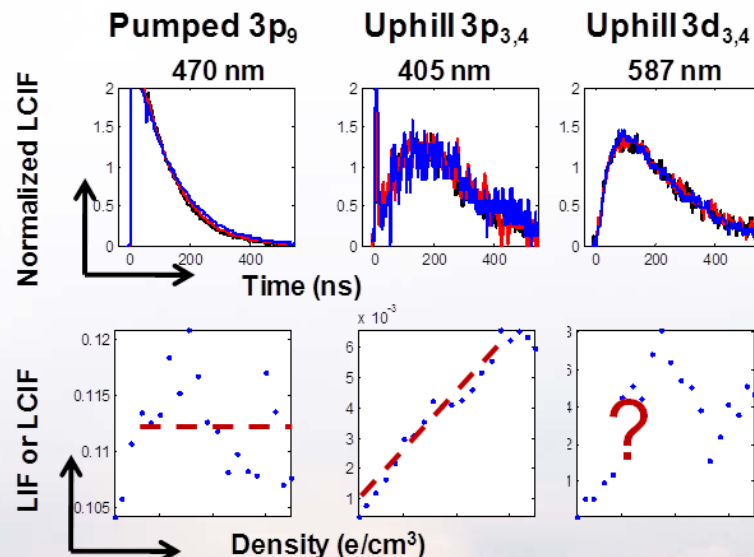
Despite reservations Argon LCIF is being investigated

- Well characterized positive columnn 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

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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 eV \sim 780 K

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

$$\Delta N_{\text{Electrons}} \sim K_{P \rightarrow D}^e n_e \Delta N_P \Delta t \quad \text{and} \quad \Delta N_{\text{Neutrals}} \sim K_{P \rightarrow D}^N n_0 \Delta N_P \Delta t$$

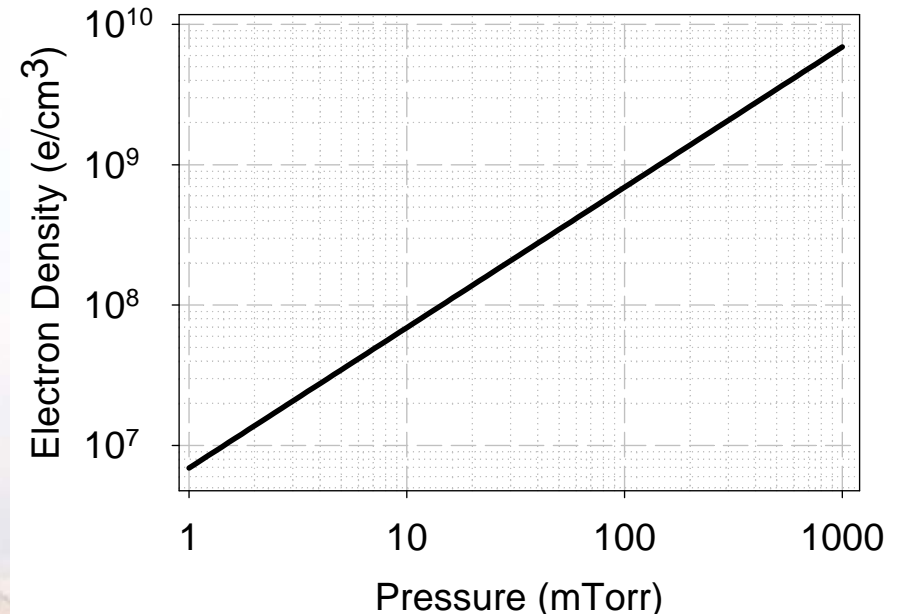
Bound determined by setting the two equal

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

Solve for n_e

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

Pressure dependent density bound



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



References for rates and cross-sections

■ Superelastic

- Klein Rosseland
- Sobelman

$$K_{ij}^e = \langle \sigma_{ij} v_e \rangle = \left(\frac{m_e}{2\pi k T_e} \right)^{3/2} \int_0^\infty \sigma_{ij}(v) \exp\left(\frac{-m_e v^2}{2k_B T_e} \right) 4\pi v^2 dv \left[\frac{g_j}{g_i} \exp\left(\frac{(E_j - E_i)}{k_B T_e} \right) \right]$$

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$$K_{ij}^e = \langle \sigma_{ij} v_e \rangle = \left(\frac{m_e}{2\pi k T_e} \right)^{3/2} \int_0^\infty \sigma_{ij}(v) \exp\left(\frac{-m_e v^2}{2k_B T_e} \right) 4\pi v^3 dv$$



Further experiments point to where improvement is needed

- Here is the "ugly" data
 - Predicted trends approach measured trends
- Two data sets offer some clues
 - Low pump power and low concentration of species
- Stimulated emission inducing 3^3P to 3^3S transition?
 - Population inversion: $N_P \gg N_S$ after excitation

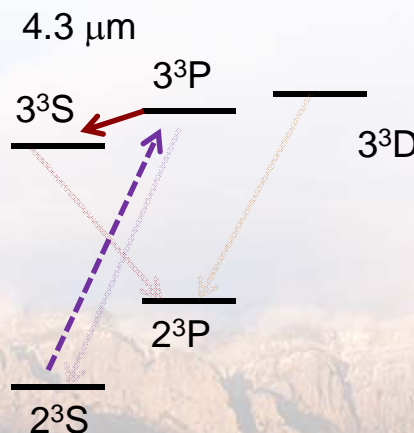
$$\frac{dN_S}{dt} = \underbrace{AN_P}_{\text{Spontaneous Emission}} + \underbrace{A\left(\frac{\lambda^2}{8\pi}\right)g(\nu)\left(N_P - \frac{g_2}{g_1}N_S\right)\frac{I}{h\nu}}_{\text{Stimulated emission \& absorption}}$$

After pumping inversion occurs

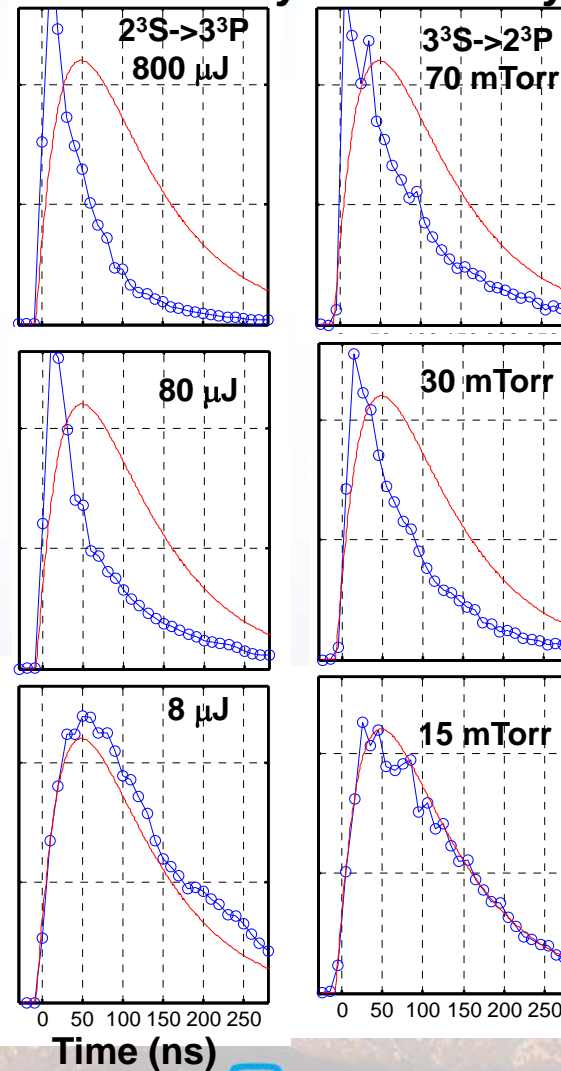
$$\frac{dN_S}{dt} \approx A_{\text{eff}}N_P, \quad N_P \gg N_S$$

Where

$$A_{\text{Eff}} = A\left[1 + \left(\frac{\lambda^2}{8\pi}\right)g(\nu)\frac{I}{h\nu}\right] > 10^8 \text{ s}^{-1}?$$



Photon density Gas density



More complete treatment should include photon densities



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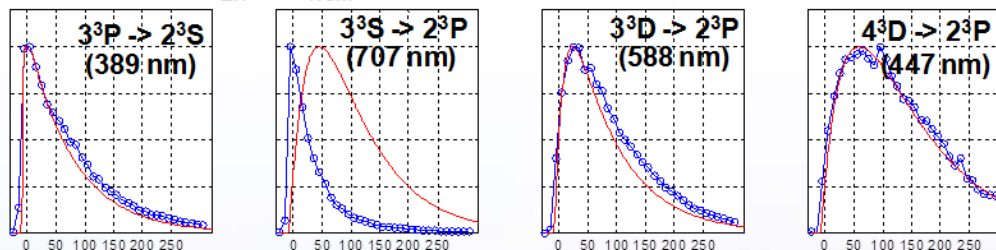
Complete treatment bypassed through simplifying approximations

- Assume population inversion occurs only during laser excitation
 - Side step need to track absolute photon intensities

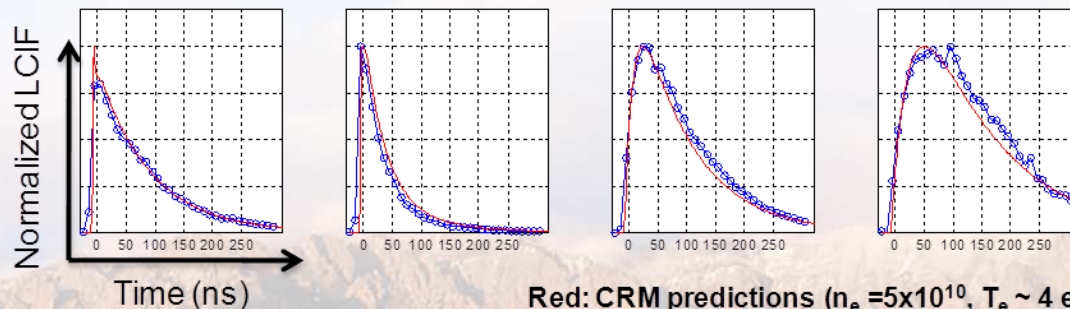
$$A_{Eff} = A_{Nom} \left[1 + A_{Inv} e^{-\frac{I_{Laser}}{I_{Threshold}}} \right]$$

Normalized time resolved trends

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



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



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

Simplifying assumption produces trends consistent with observation



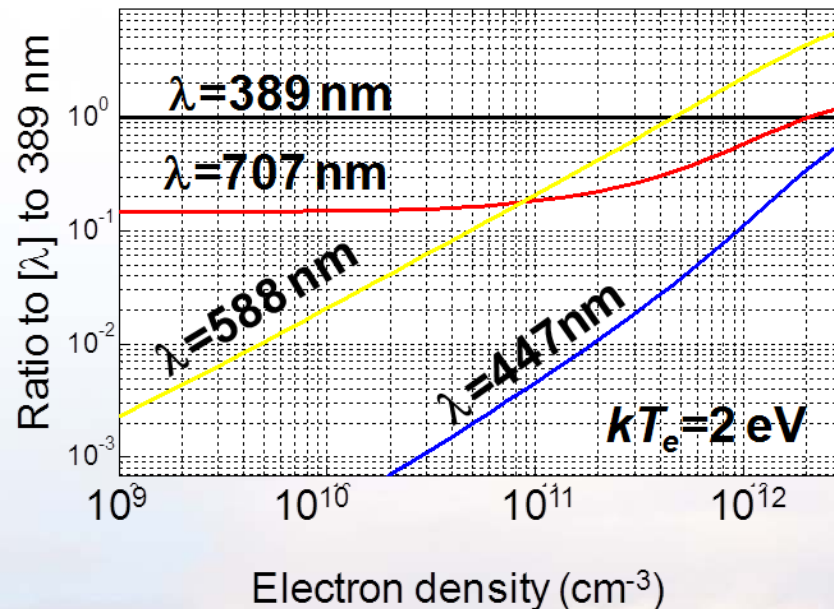


Key uphill transitions are not significantly impacted by radioactive coupling

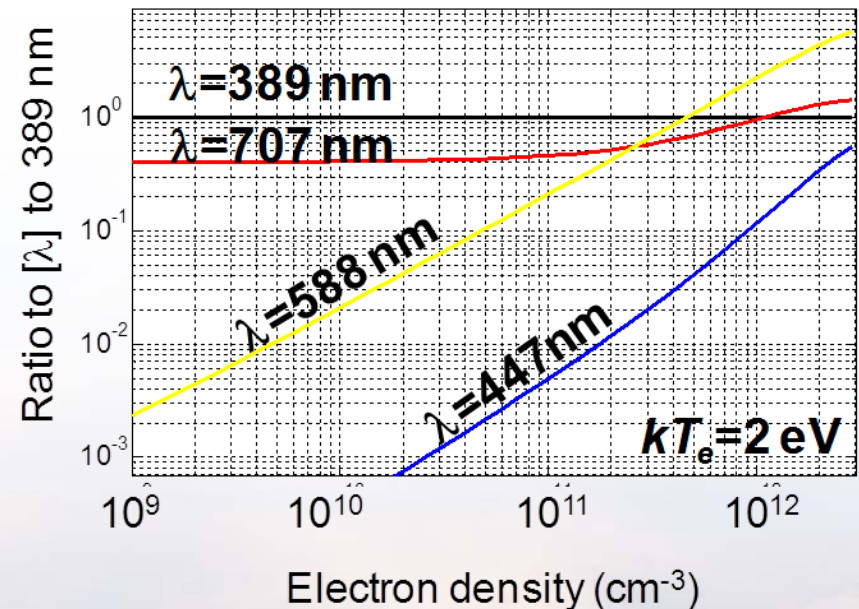
- Dominant population pathway is still through excited 3^3P state
 - Final densities of 3^3P state will change, but this is normalized out in analysis

Averaged trends

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



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

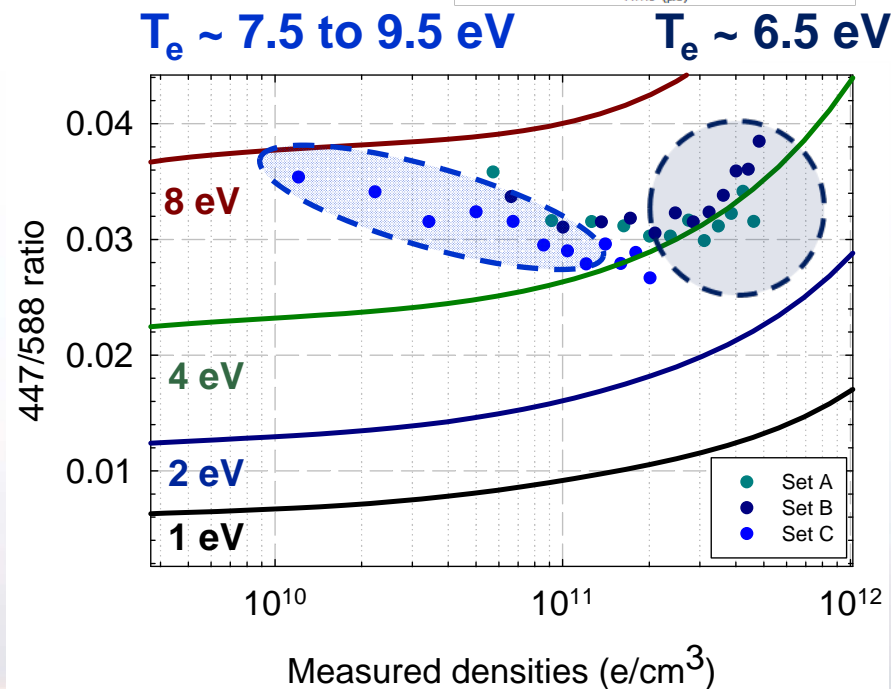
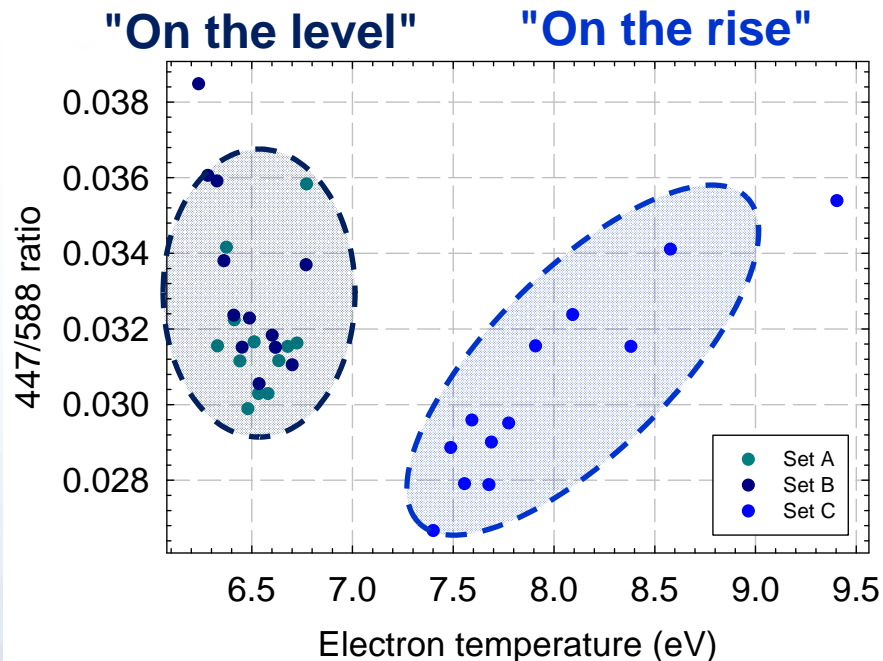
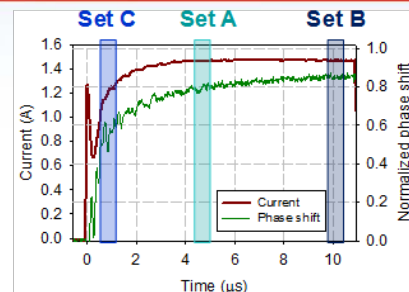


Caution should still be used if considering the 3^3S state for normalization



[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

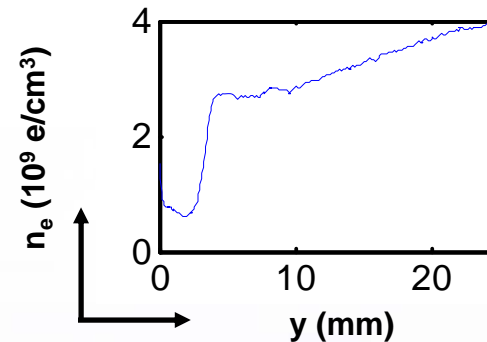
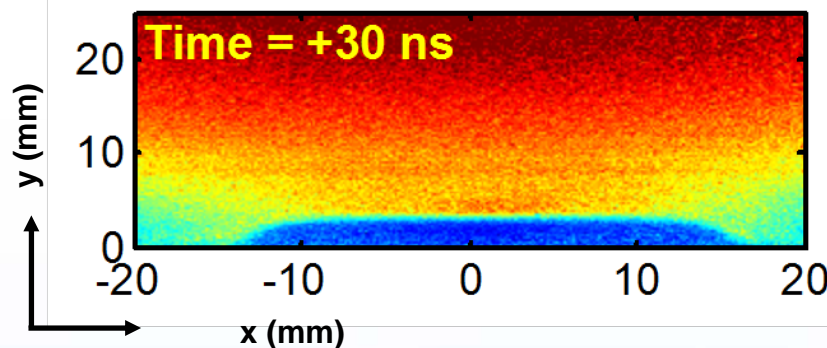


Uncertainties in rates, EEDF and/or interpolation of T_e from drift parameters should impact absolute values



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:

$$\nabla^2 V = \frac{e}{\epsilon_0} n \longrightarrow n_i \approx \frac{2\epsilon_0 V}{e\Delta x^2}$$

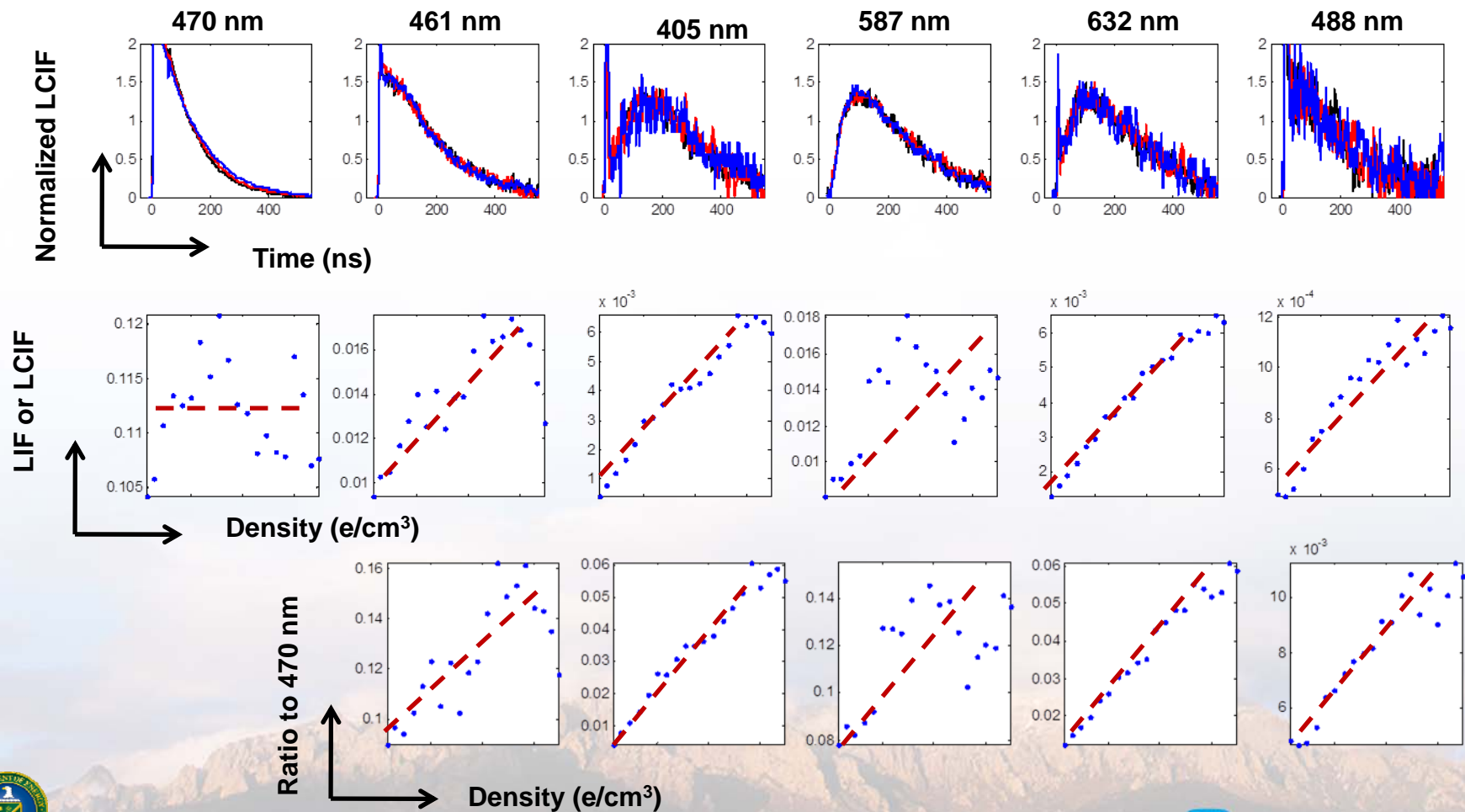
- For a voltage of 1 kV and a thickness of ~ 5 mm:

$$n_e \sim 4 \times 10^9 \text{ electrons/cm}^3$$

Reasonable cross-check of LCIF technique

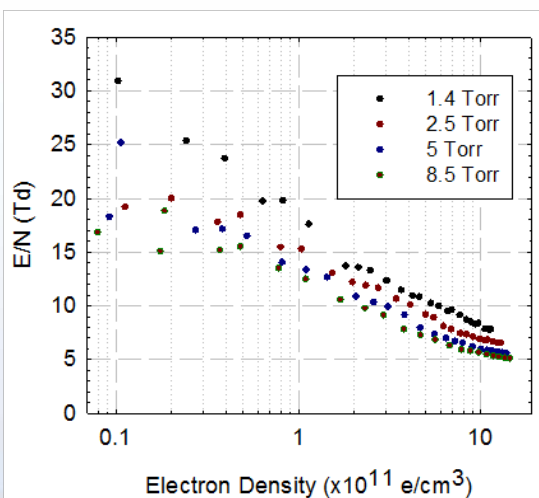


Preliminary survey points to useful transitions

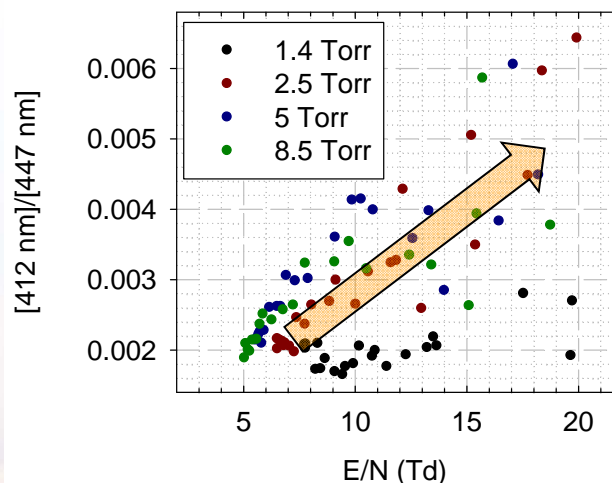


Preliminary investigation of proposed scheme looks promising

Extracted E/N



[412 nm]/[447 nm]



[403 nm]/[447 nm]

