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Time resolved evolution of transient plasmas as measured with laser-collision induced fluorescence

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LOCKHEED MARTIN

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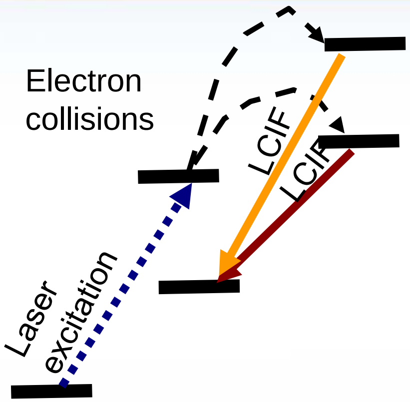
LCIF provides 2D snapshots of plasma density and temperature

- **Motivation: What is the density? What is the temperature? Where and When?**
  - Electrical probe based 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
    - Describe LCIF technique and application to triplet manifold of Helium
    - Experimental implementation of LCIF
  - Applications of LCIF
    - Pulsed plasmas
    - Transient anodic plasma structure
    - Ion sheath expansion and collapse
  - Future directions and concluding thoughts

LCIF technique relies on redistribution of excited states by (electron) collisions

- **This is LCIF**
  - Electrons redistribute laser-excited population
  - Monitor changes in emission from coupled states
- **Employ a collisional-radiative model (CRM) to predict redistribution**
  - Utilize limited basis of states to reduce number of equations
  - Helium, 15 states (ground + 14 triplet states)



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

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

- **Electron density and electron temperature appear in first term**
  - Temperature dependence introduced via K^e_{ij} t

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

- **Technique has been employed for over 30 years**

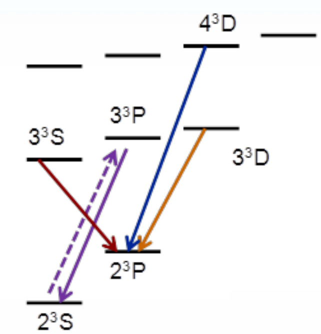
We extend the technique for use in two-dimensions and obtain time resolved information

Ratios of intensity are utilized for determining n_e, kT_e

- **Utilizing ratio of intensities**
  - Eliminate problems associated with absolute calibration

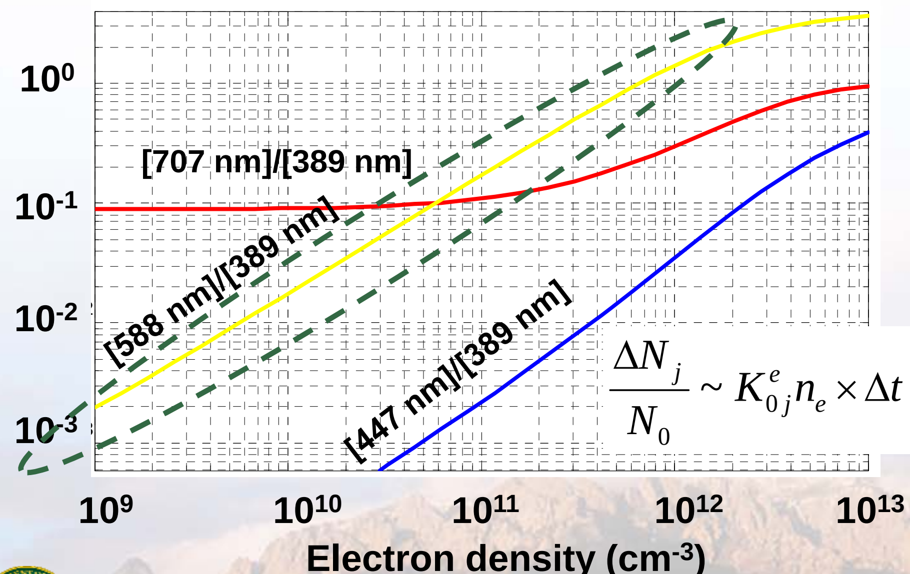
$$\Delta N_j \sim K^e_{0j} n_e \times N_0 \times \Delta t \quad \Rightarrow \quad \frac{\Delta N_j}{N_0} \sim K^e_{0j} n_e \times \Delta t$$

Key transitions

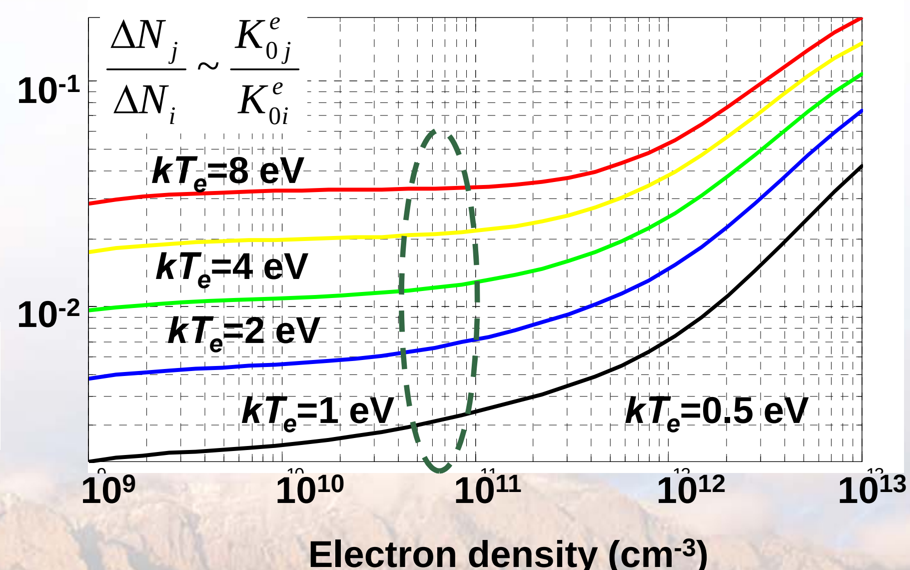


- **Capitalize on weak kT_e dependence of $3^3P \to 3^3D$**
  - Ratio of 588 nm to 389 nm yields n_e;
  - Ratio of 447nm to 588 nm yields kT_e

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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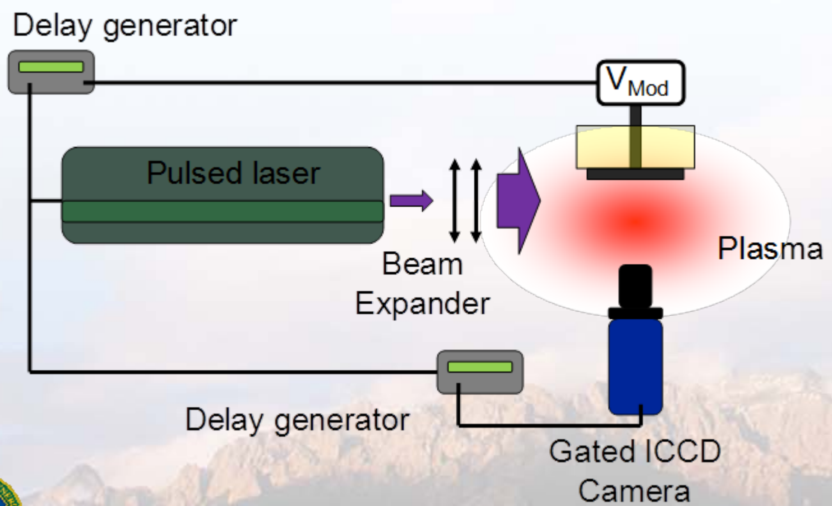
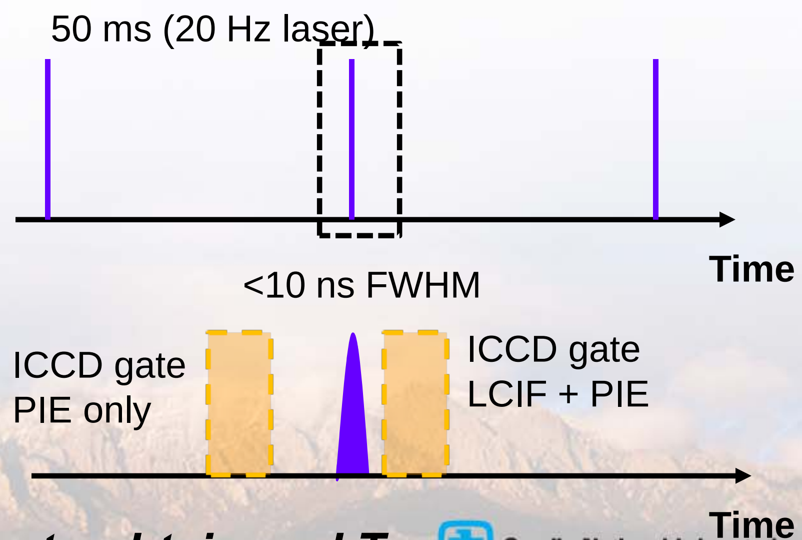
Experimental implementation of the LCIF is realized

- **Nanosecond pulsed laser used for excitation**
  - < 10 ns FWHM @ 20 Hz
- **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

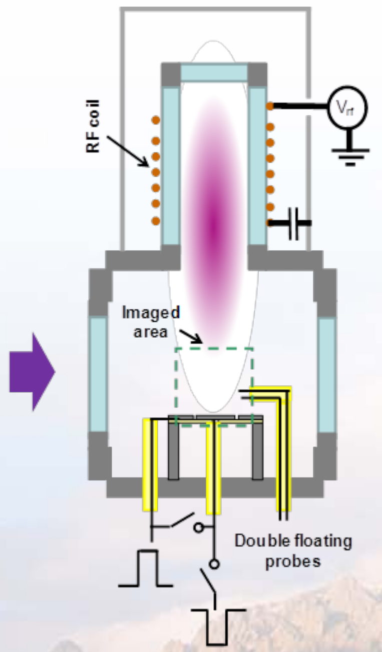


Need to make six (3 x 2) measurements to obtain n_e, kT_e

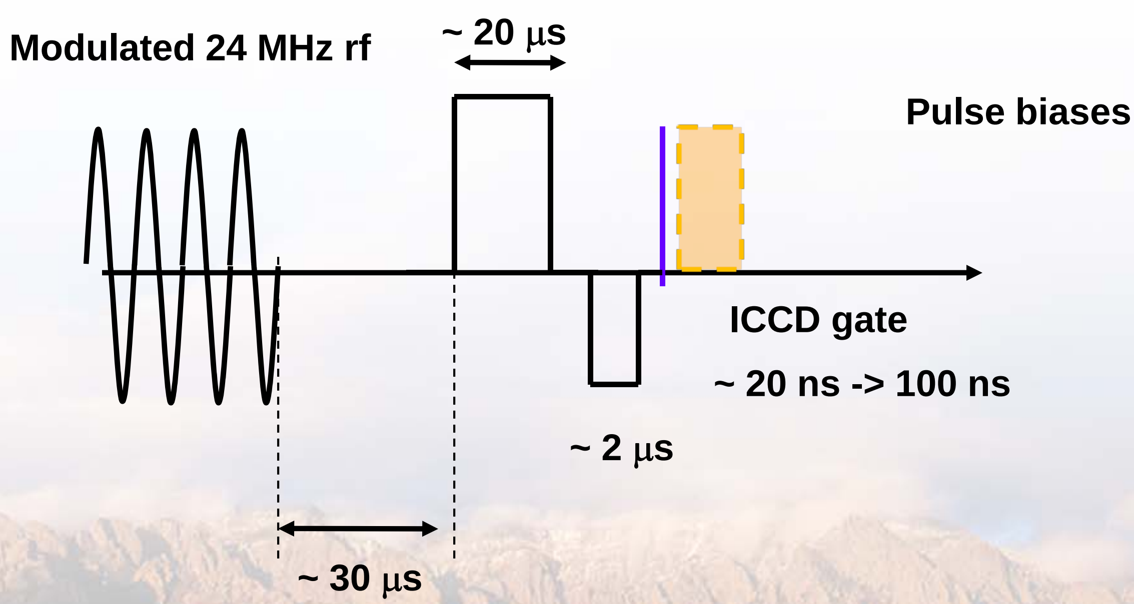
Experiment designed for flexibility

- **Time modulated rf plasma**
  - Generate metastable "seed " to prepare for transient measurements
- **Segmented electrodes**
  - Positive and or negative polarity pulses
- **Computer controlled delays**
  - Time step across event of interest

Setup



Timing sequence



Examine phenomena on different spatial and temporal scales

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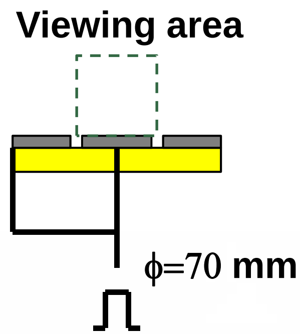
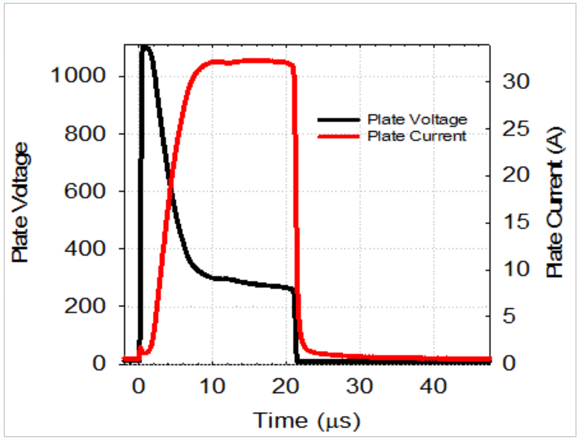
Plasma generated by time modulated anode tests LCIF technique

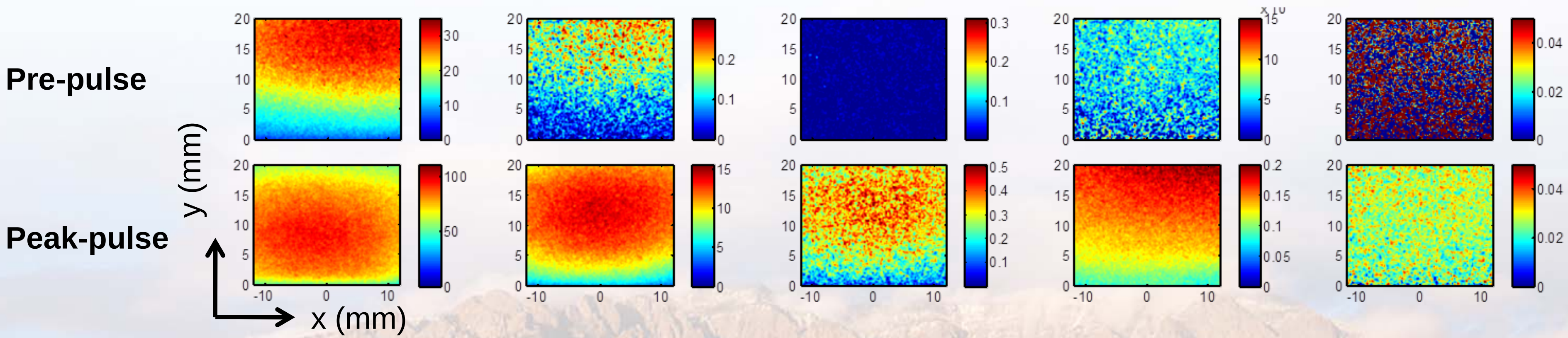
- **Challenging plasma system**
  - Dynamic plasma potential
- **Examine time evolution of transient plasma**
  - RF afterglow, 50 mTorr He
  - Positive pulse bias (~ 1 kV) applied to planar electrode

Setup and behavior





Representative LCIF Images and ratios



Two dimensional images vary by orders of magnitude in intensity

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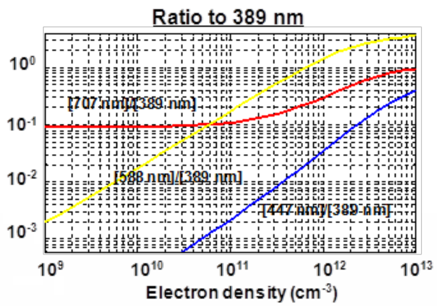
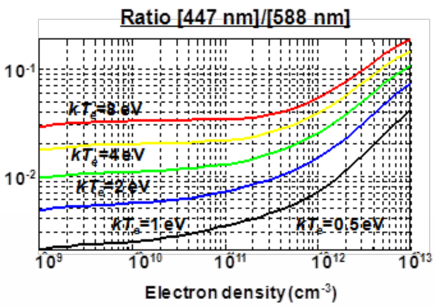
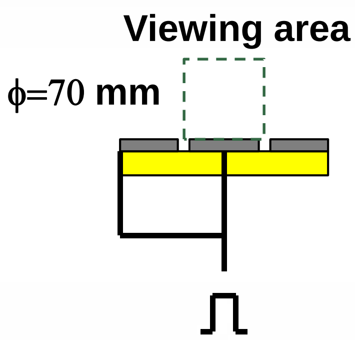
LCIF data analyzed with CRM predictions to yield n_e, kT_e

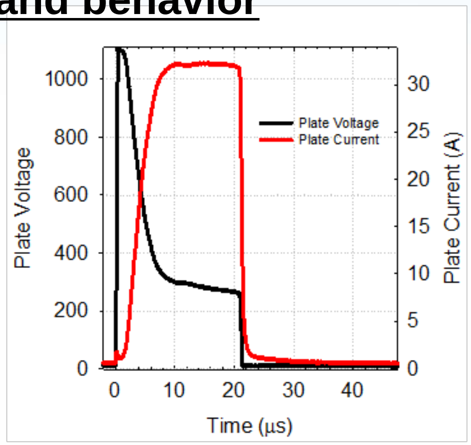
- **Trends analyzed with CR model**
  - Produce n_e, T_e as functions of time



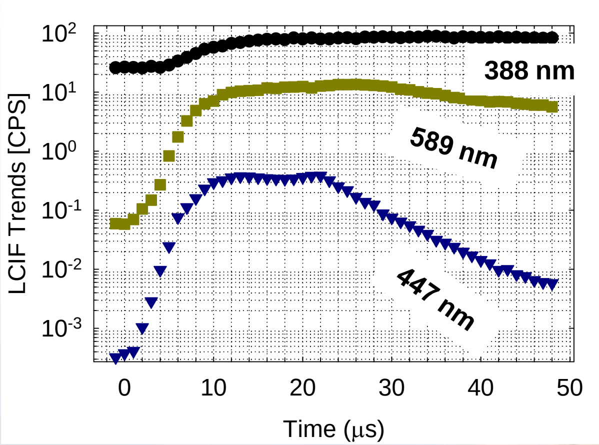


Setup and behavior

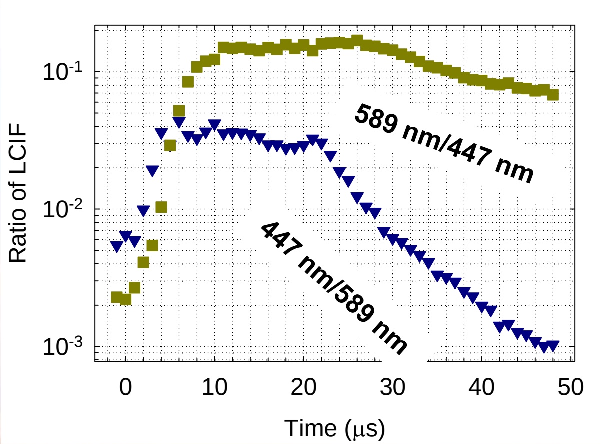




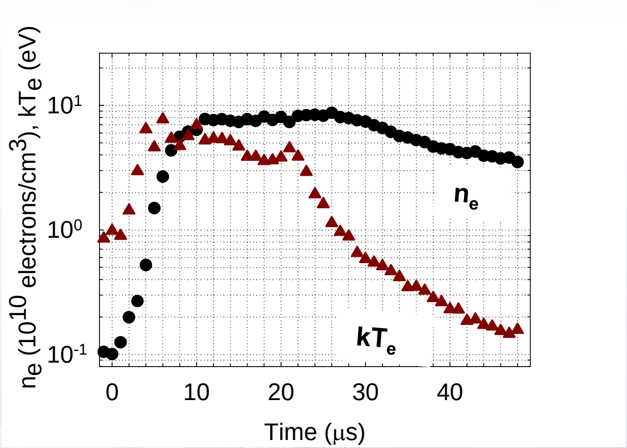
LCIF Trends



LCIF Ratios



Densities and temperatures



Densities and temperatures vary by orders of magnitude; rates demonstrate different time scales

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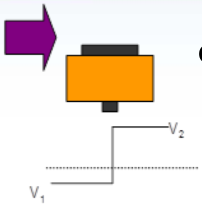
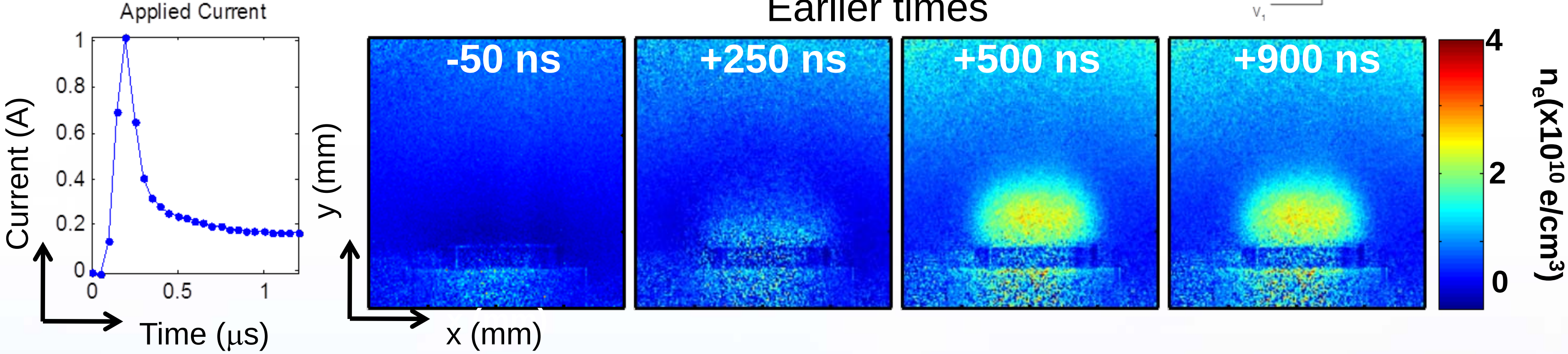
Transient structure of plasma observed after pulse excitation

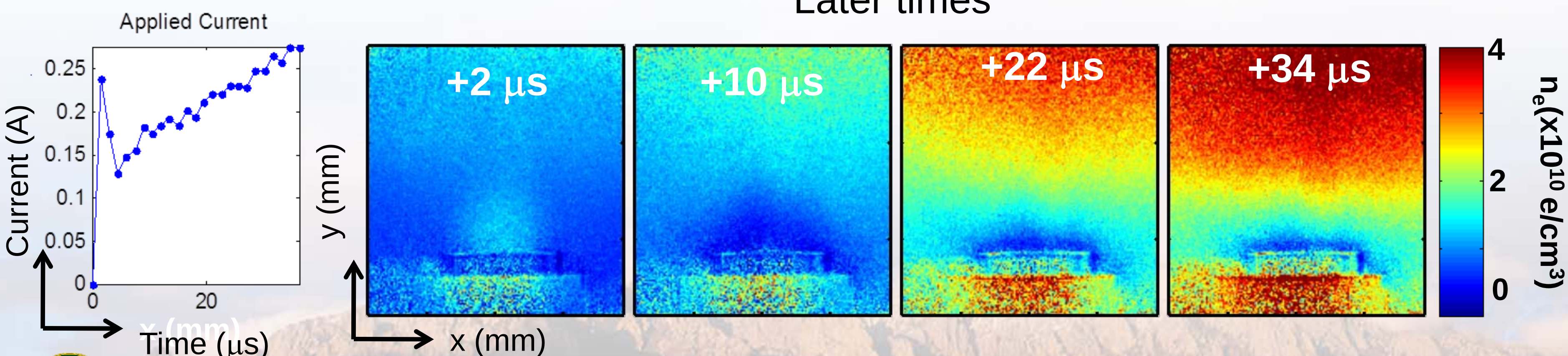
- **Closer analysis of initial plasma distribution**
  - Use smaller (25 mm) diameter electrode, 100 mTorr afterglow



Earlier times



Later times



Initial plasma distribution is unstable

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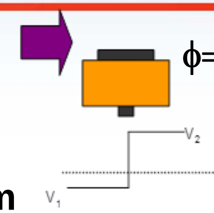
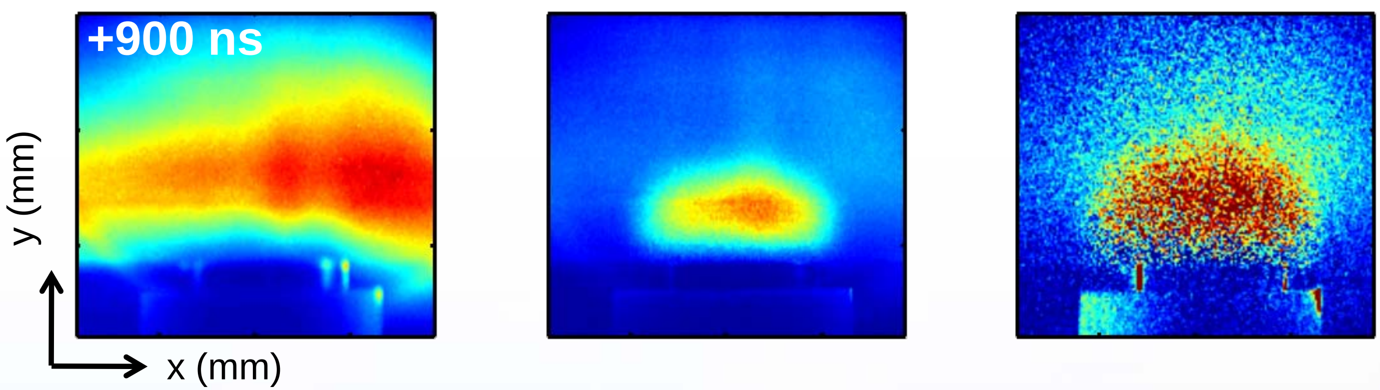
Higher energy electrons observed around edge of anode plasma

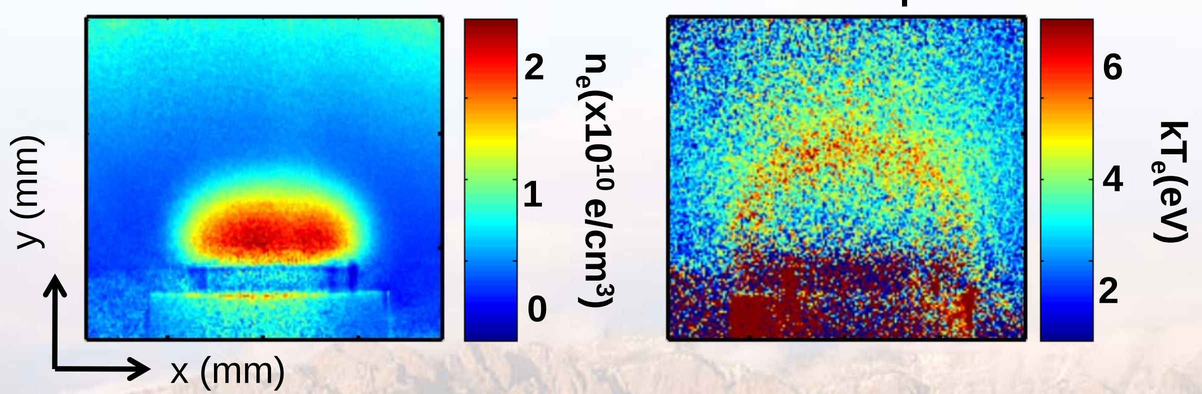
- **Temperature measurements made for +900 ns case**
  - Challenging measurement because of low level signals



LCIF Data



Analysis



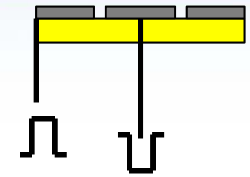
"Anodic Fireball"

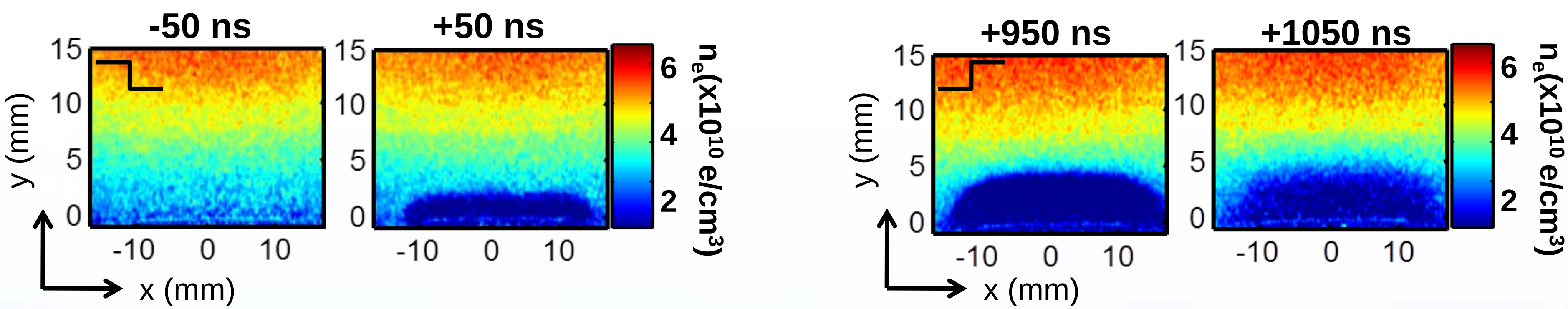
Electrons energized by localized electric fields supporting double layer

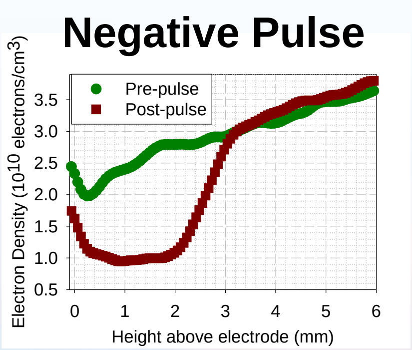
S. D. Baalrud , B. Longmier and N Hershkowitz, Plasma sourc. Sci. Technol. 18 035002 (2009)

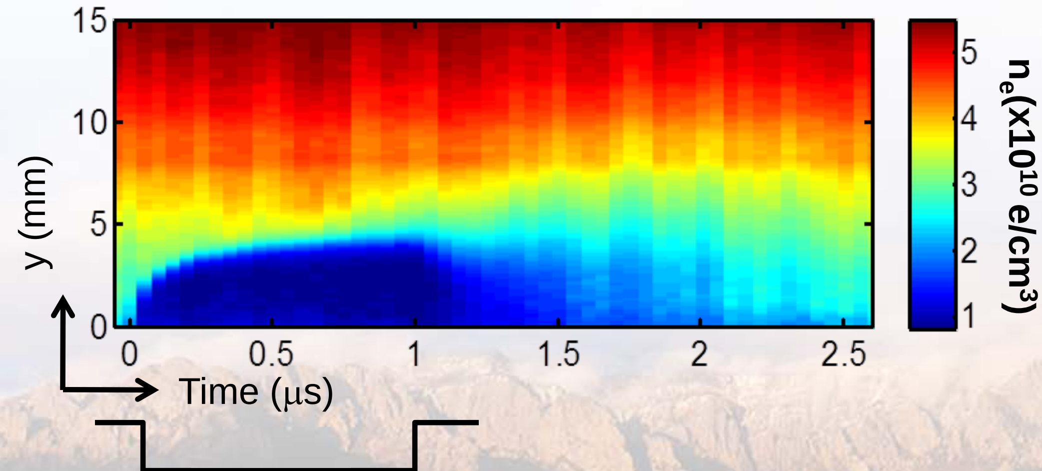
LCIF captures 2D-sheath formation

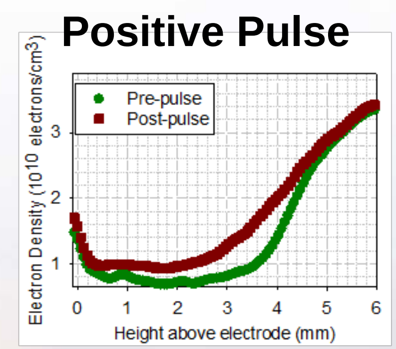
- **Examine evolution and structure of ion sheath**
  - - 1 kV, 1 μs bias applied to inner electrode, 5 μs after + pulse ends
  - 20 ns snapshots of LCIF









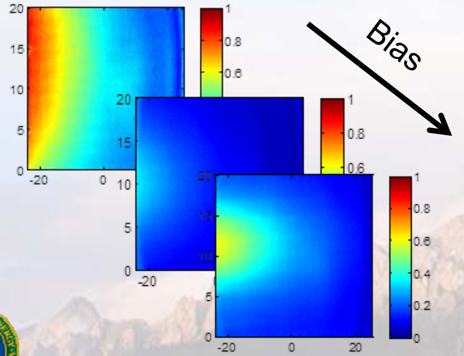


Transient sheath dynamics captured by LCIF

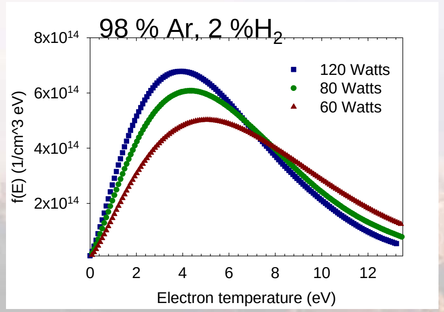
Concluding remarks and future directions

- **LCIF technique demonstrated in 2D**
  - Good spatial resolution – limited by optical collection
  - Decent temporal resolution – limited by ICCD gate times & tolerable signals
- **Technique should be extendable over broad parameter space**
  - Higher pressures – neutral collisions
  - Other atomic systems - Argon
- **Looking into**
  - ECR based plasma systems (B. Weatherford and J Foster, U. Michigan)
  - Rare gas/Hydrogen mixtures (A. El Saghir and S. Shannon, NC State)
  - Fast Ionization wave (W. Lempert and I. Adamovich, Ohio State)

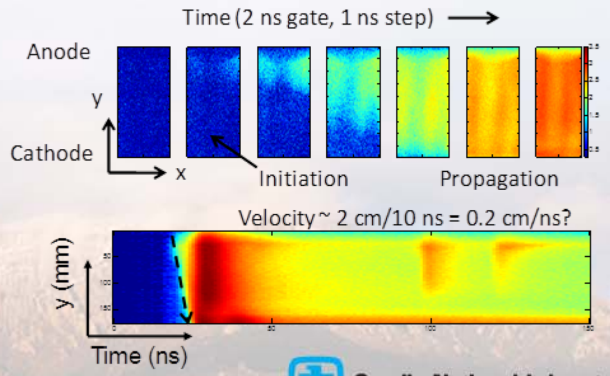
ECR Plasma



RG/H_2 EEDF



FIW



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Thank you!

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Contract DE-SC0001939

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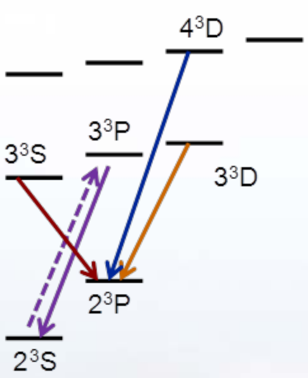
Helium atom serves as target species for LCIF measurements

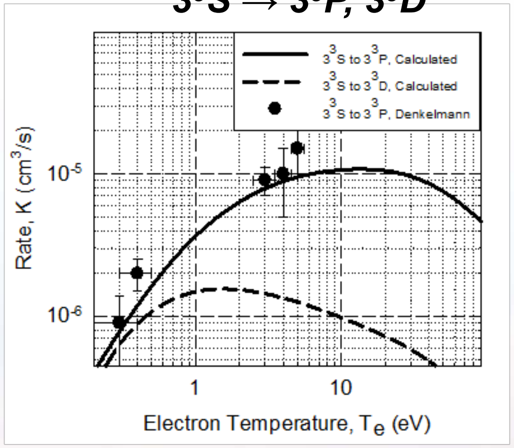
- **Employ Helium to start with**
  - "Simple system" with "better known" rates
- **Utilize functionalized form of cross-sections compiled by Ralchenko[1]**
  - Integrate to get rates, compare to measured rates [2,3]

Key transitions

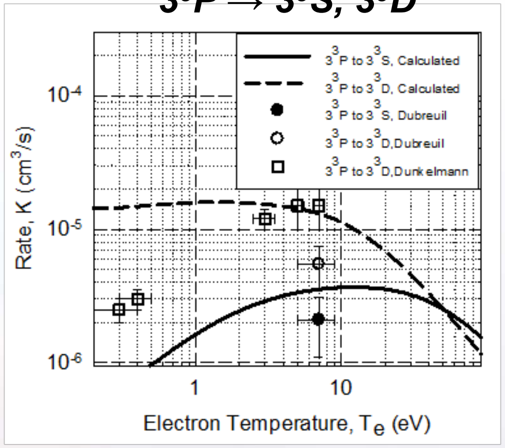


Computed and measured excitation rates in Helium

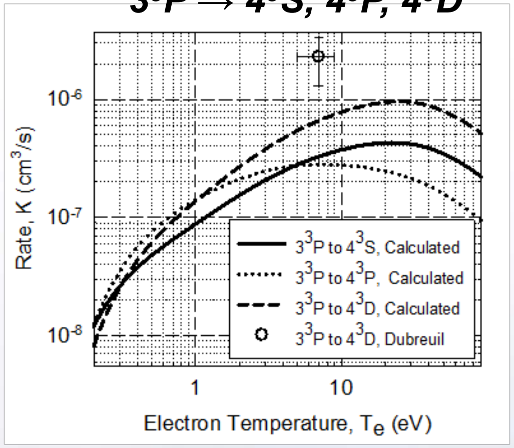
$3^3S \rightarrow 3^3P, 3^3D$



$3^3P \rightarrow 3^3S, 3^3D$



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



1: 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)
2: 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).
3: 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 $K_{ij}(kT_e)$

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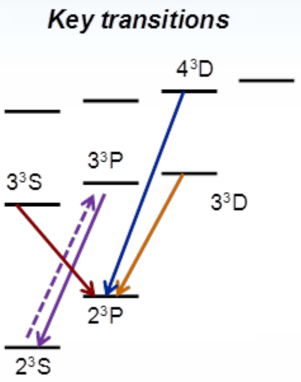
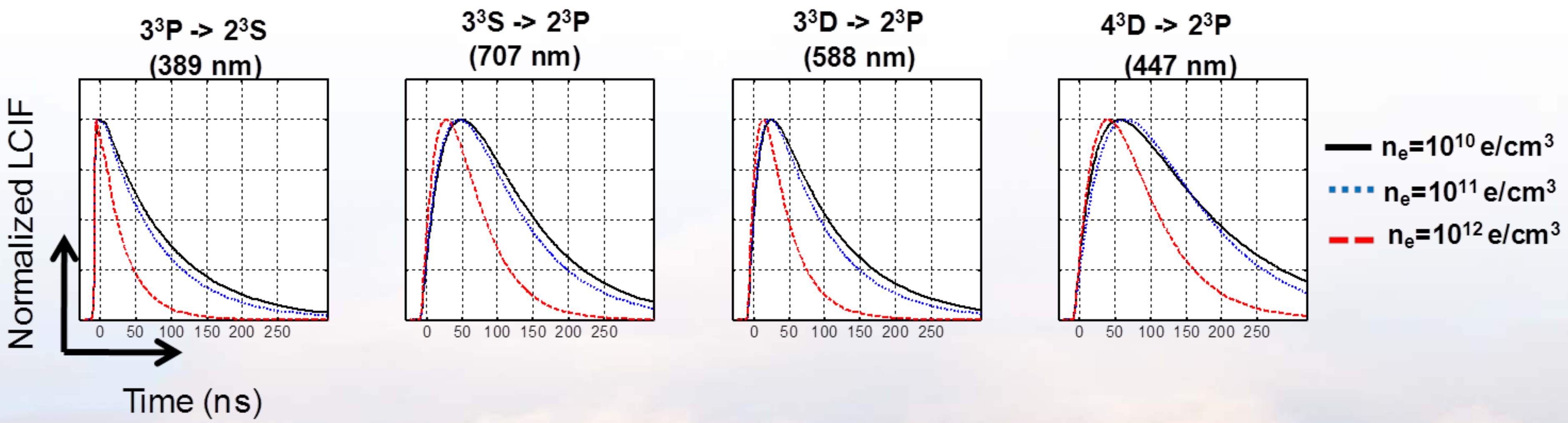
CRM predicts evolution of various Helium states after laser excitation

- **Computed evolution of LCIF**
  - After laser excitation of 3^3P from 2^3S
- **Temporal evolution may serve as "fingerprint" of electron interaction**
  - Below $n_e \sim 10^{11}$ absolute intensities are needed
  - Analyze shape of decay above $n_e \sim 10^{11}$ electrons/cm^3



<u>Computed temporal evolution</u>



Need at least two time-resolved profiles to uniquely obtain n_e, kT_e

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References for rates and cross-sections

- **Superelastic**
  - Klein Rosseland
  - Sobelman

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

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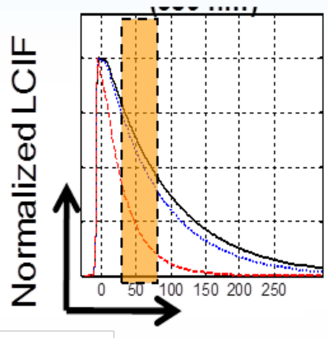
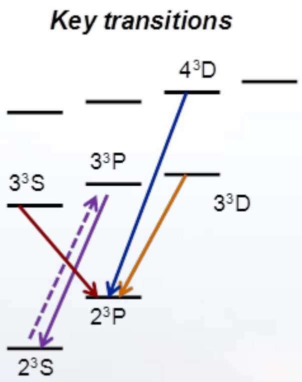
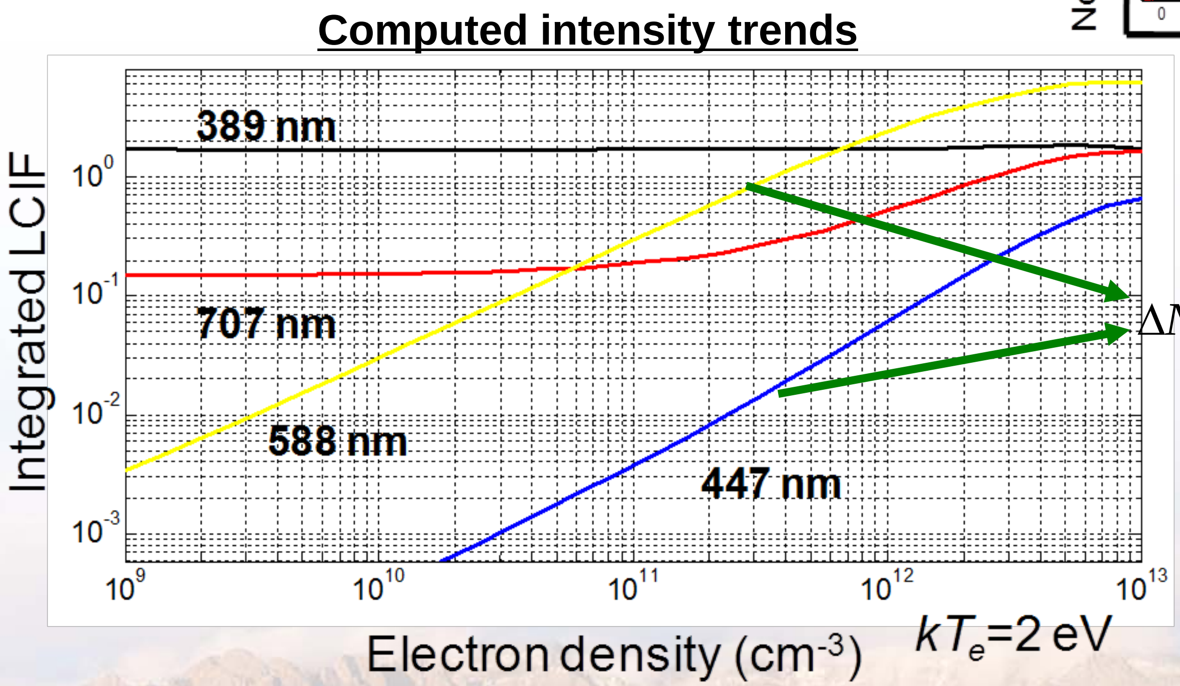
[1] I.I. Sobelman, L. A. Vainshtein, and E. A. Yukov, *Excitation of Atoms and Broadening of Spectral Lines* (Springer, New York, 1981) p. 5.

Two ways of determining density and temperature

- **Time resolved measurements involve measuring decay of LCIF**
  - ~ 100 ns, multiple temporal points to resolve shape
- **Integrate LCIF over some defined temporal window**
  - Short integration times, better temporal resolution







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

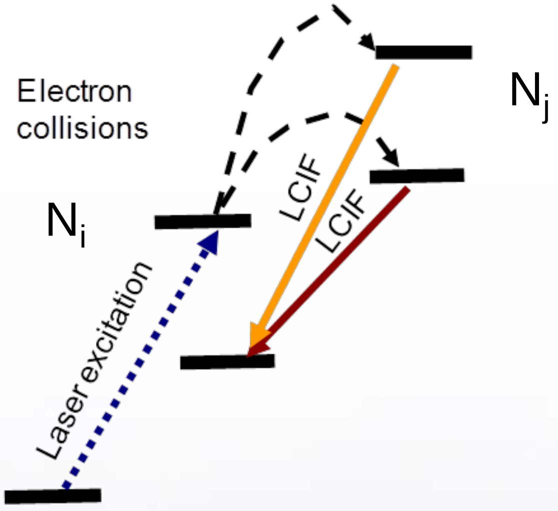
Intensity of LCIF from 3^3D and 4^3D exhibit linearity over 3 orders of magnitude, LIF from 3^3P flat

Simplification lends insight into what technique measures

- **Reduce coupled sets of equations to emphasize key point**
  - Consider population of "uphill" transitions

$$\frac{dN_j}{dt} \sim K_{ij}^{e} n_e \times N_i$$

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



LCIF response is convolution of n_e and $K(kT_e)$, which is generally a complex function of kT_e

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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: $N_P >> N_S$ after excitation

Spontaneous Emission ***Stimulated emission & absorption***

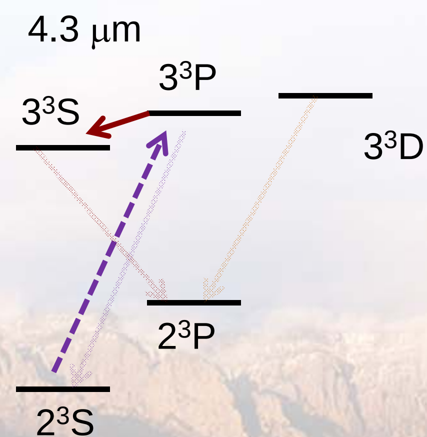
$$\frac{dN_S}{dt} = AN_P + A\left(\frac{\lambda^2}{8\pi}\right)g(\nu)\left(N_P - \frac{g_2}{g_1}N_S\right)\frac{I}{h\upsilon}$$

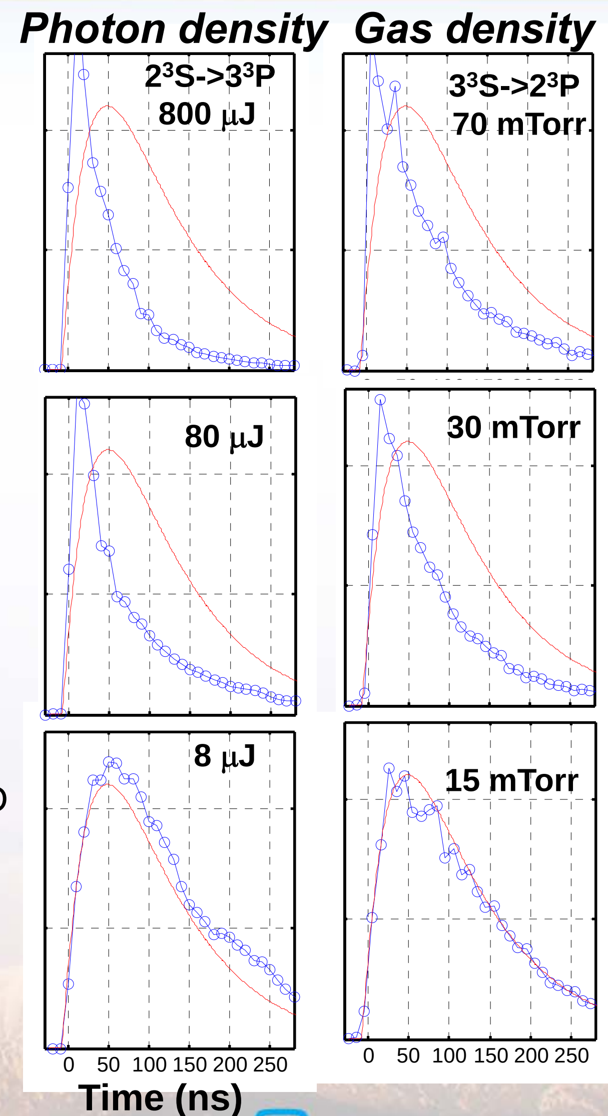
After pumping

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

Where

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



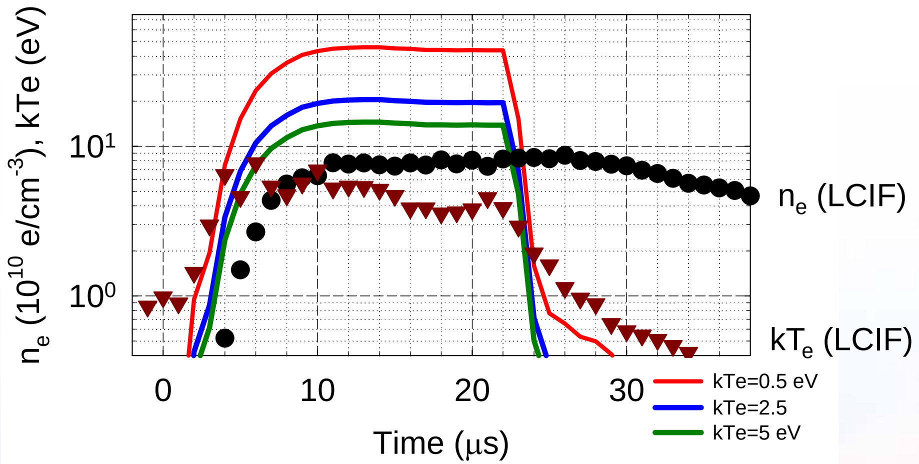


More complete treatment should include photon densities

Analysis of pulsed plasma offers second means to benchmark LCIF technique

- **Further test measurements made by LCIF**
  - Two approaches to quantify densities and temperatures
  - Both have their own "baggage"

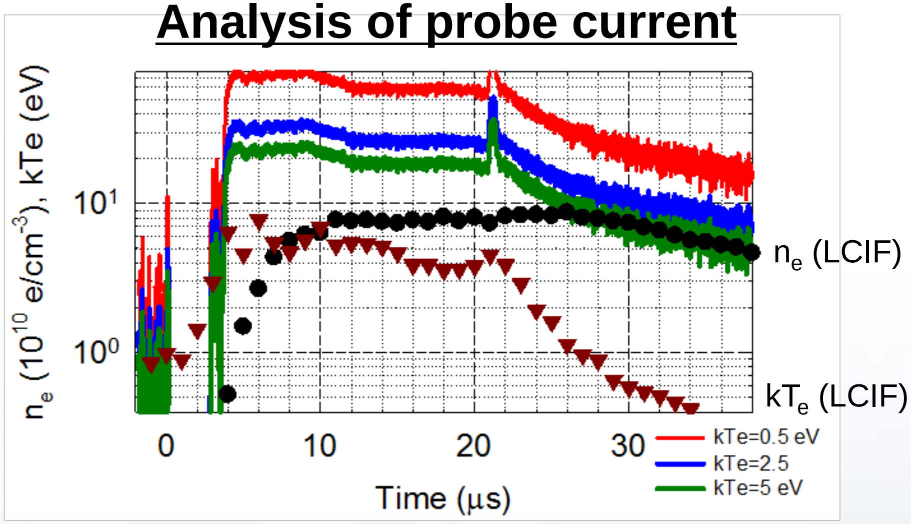
Analysis of plate current



$$I_{Plate} = \tfrac{1}{2} n_e u_e A_{Plate} e^{\frac{(V_{Plate} - V_{Plasma})}{kT_e}}$$

$$I_{Plate} \approx 0.18 n_e u_e A_{Plate} \quad (A_{Chamber}/A_{Plate} \sim 50)$$

Analysis of probe current



$$I_{Probe} = n_e u_{Bohm} A_{Effective}$$

$$I_{Probe} \approx n_e u_{Bohm} A_{Probe}$$

Trends again appear "reasonable", density seems systematically low by a factor of 3….

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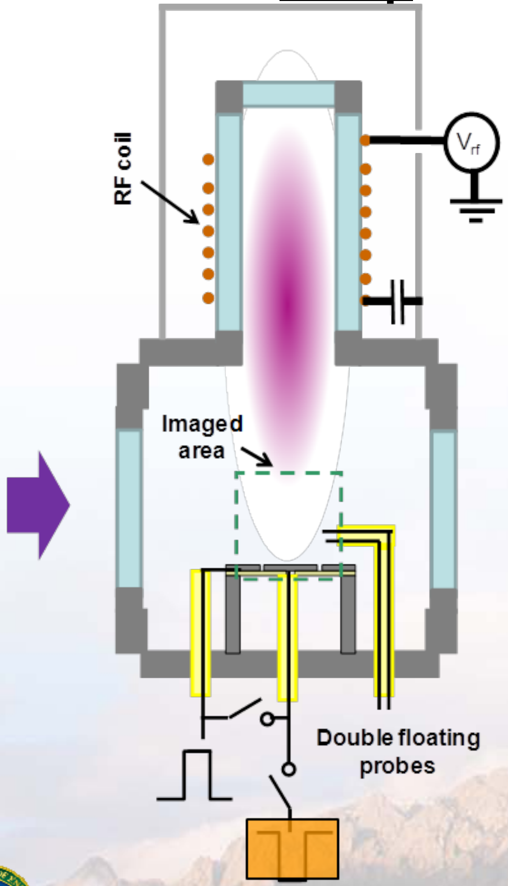
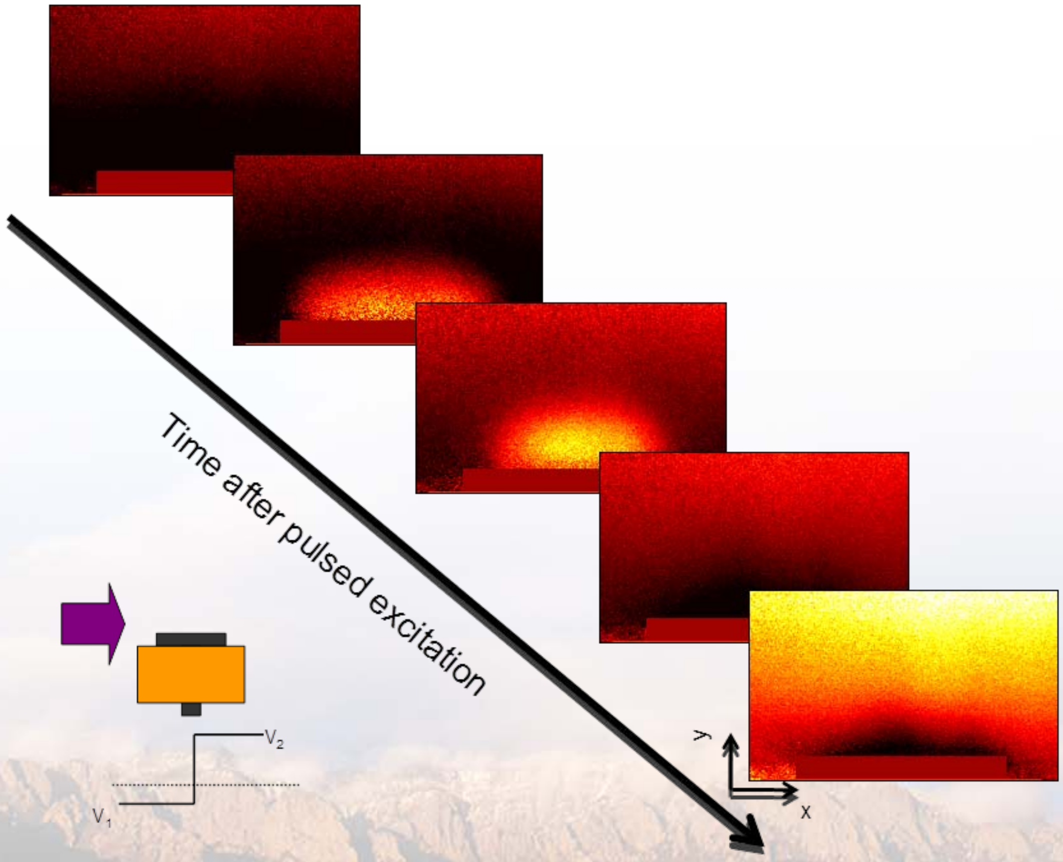
Demonstration of LCIF technique: Transient double layers

- **Bias electrode positive with respect to chamber**
  - 1 kV bias applied to inner electrode, 50 μs into afterglow (low n_e, low kT_e)

Setup



Data

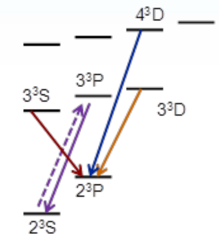


Clearly separated plasma layers

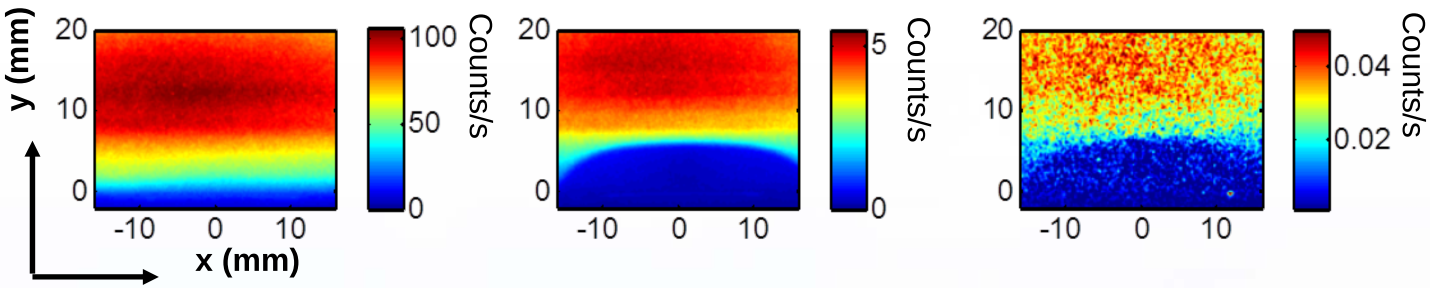
Static structure of a sheath is measured in Helium afterglow

- **Examine spatial structure around biased electrode**
  - Representative LCIF data used for analysis
  - ~ 5 microseconds into 20 mTorr Helium afterglow
  - 25 mm diameter electrode, biased to - 1 kV.

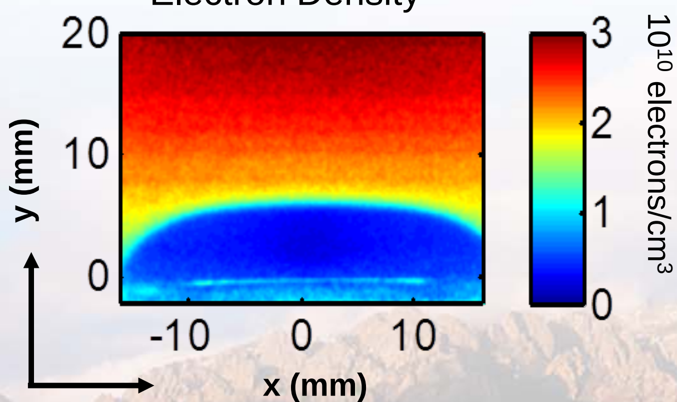
Key transitions



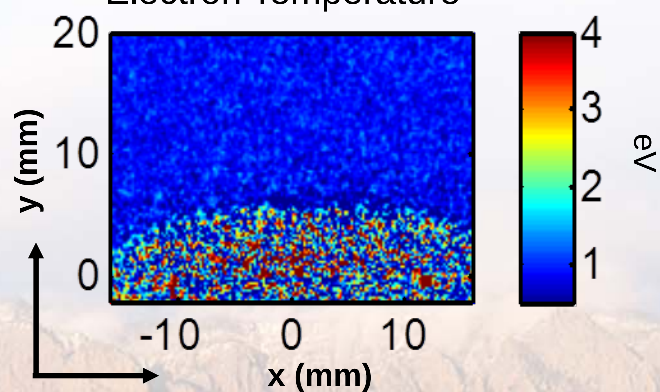
3^3P LIF (@389 nm) 3^3D LCIF (@588 nm) 4^3D LCIF (@447 nm)



Electron Density



Electron Temperature



Temperature measurements become questionable in the sheath….

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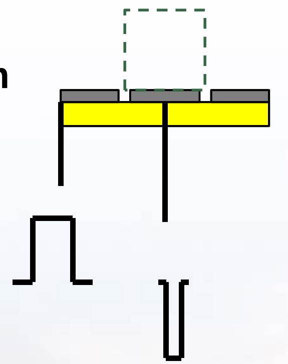
LCIF captures 2D-sheath formation

- **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

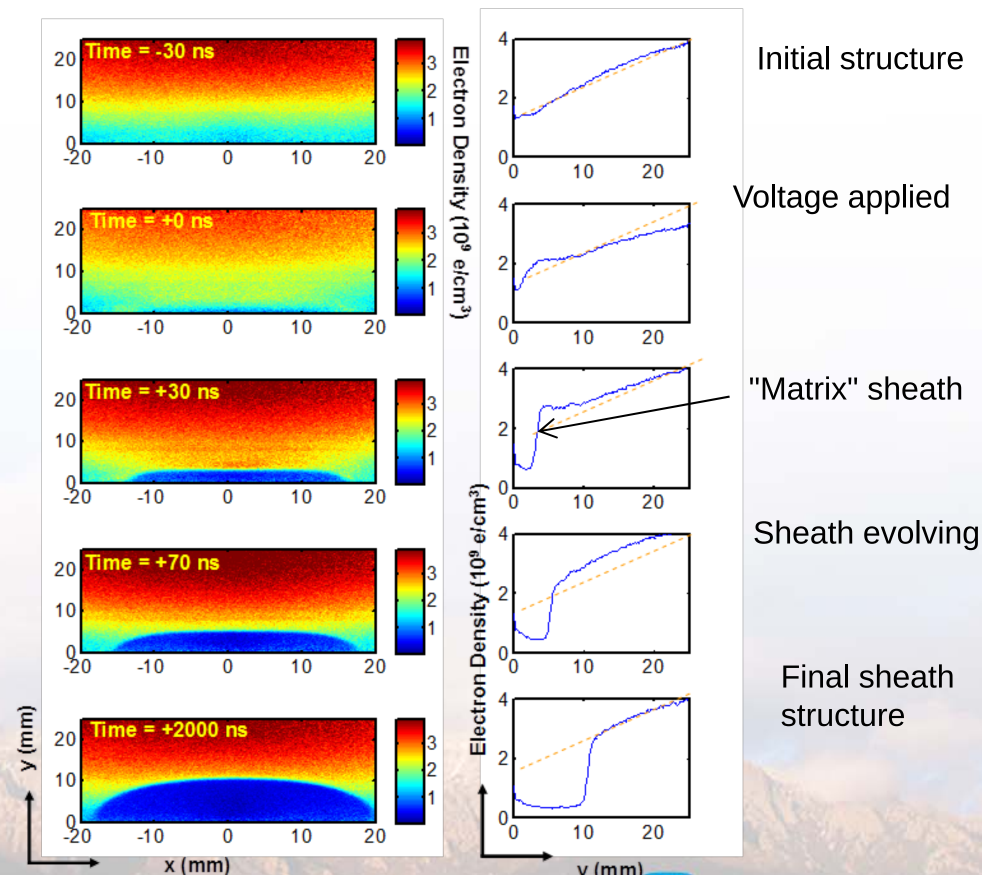
Setup

Viewing area

φ=70 **mm**



Data



Decent temporal and spatial resolution demonstrated

LCIF captures 2D-sheath formation

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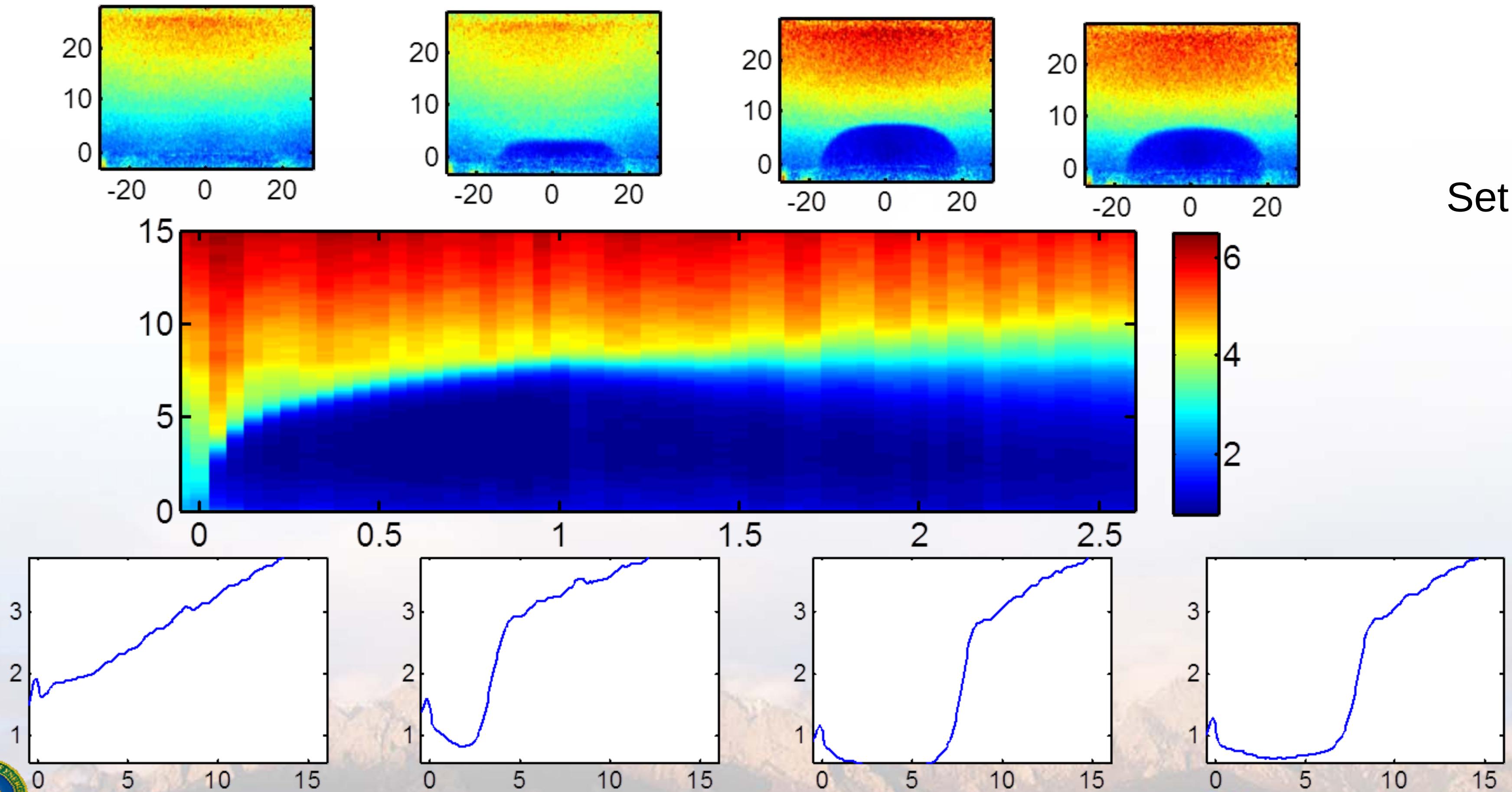

Set 6

Decent temporal and spatial resolution demonstrated

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