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IONIZATION WAVE DYNAMICS OF A PLASMA JET IN CONTACT WITH LIQUID WATER*

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AGENDA

- Plasma jets onto Liquids
- Description of Experiment
- Laser Collisional Induced Fluorescence (LCIF)
- Plasma Jet Contacting TiO_2 (ε_r = 80)
- Plasma Jet Contacting Liquid Water
- Modeling and Future Work

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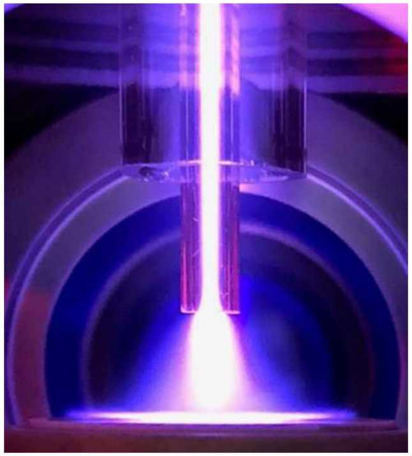
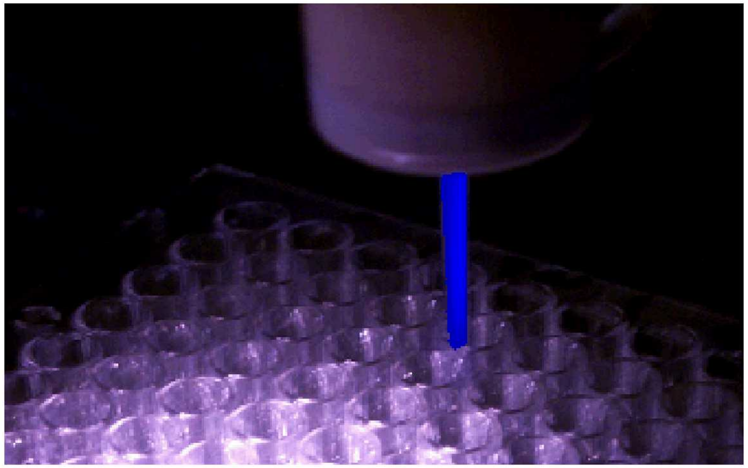
ATMOSPHERIC PRESSURE PLASMA JETS

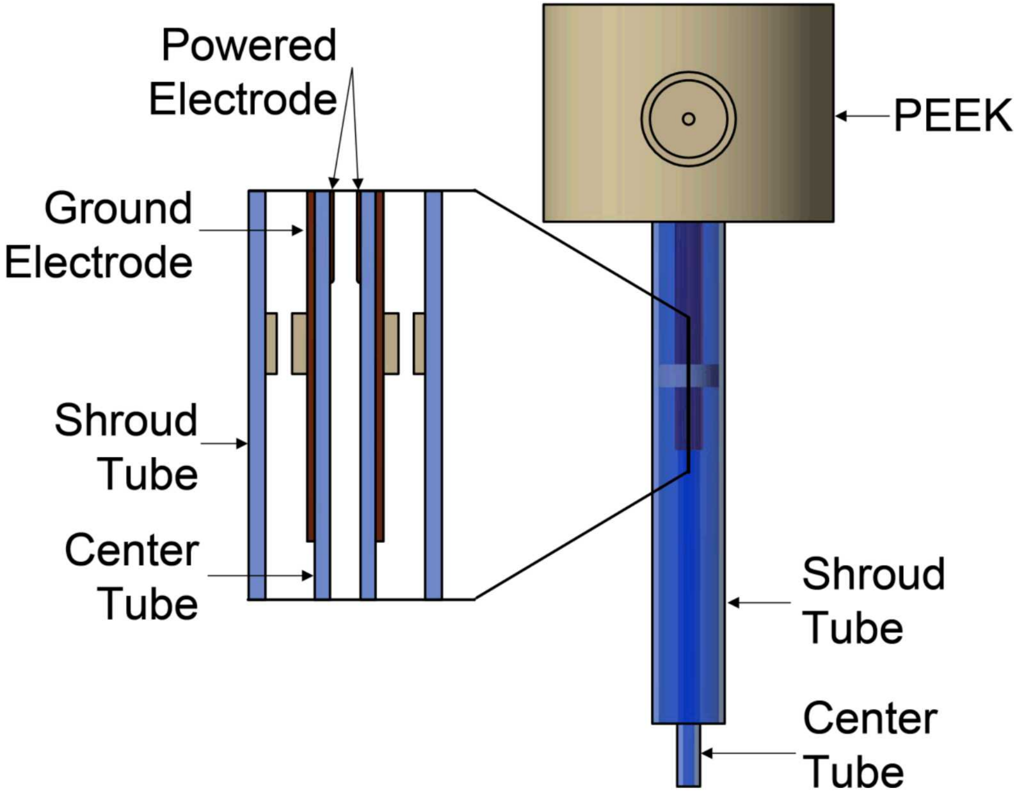
- **Atmospheric pressure plasma jets (APPJs) are a popular source of chemistry for biomedical applications.**
- **The plasma propagates as an ionization wave (IW) that is repetitively pulsed.**
- **The IW gives rise to reactive oxygen and nitrogen species (RONS) which produce the biological effect.**
- **Objective: Investigation IW dynamics in a plasma jet contacting liquid in a well controlled environment.**

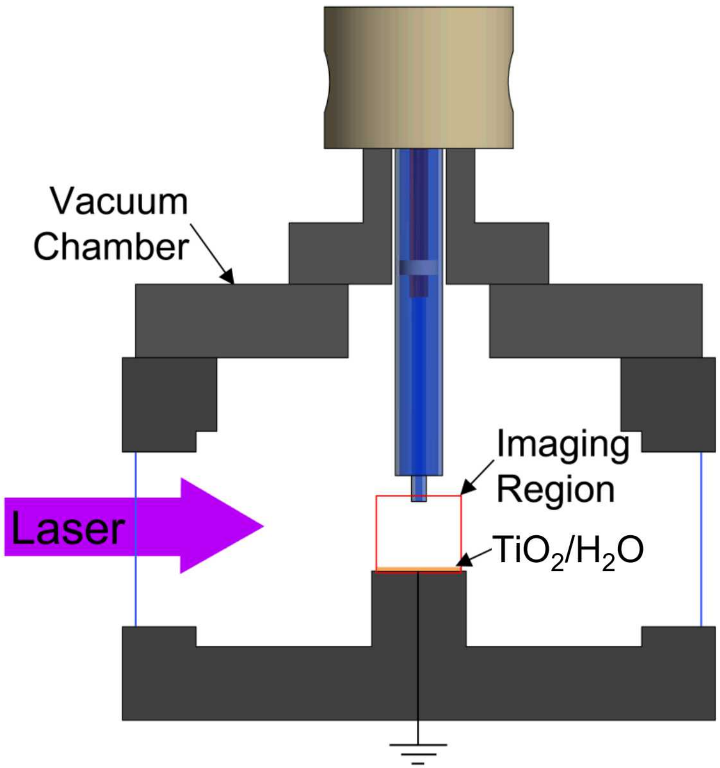




- S. Mohades, et al., Physics of Plasmas 22, 122001 (2015).

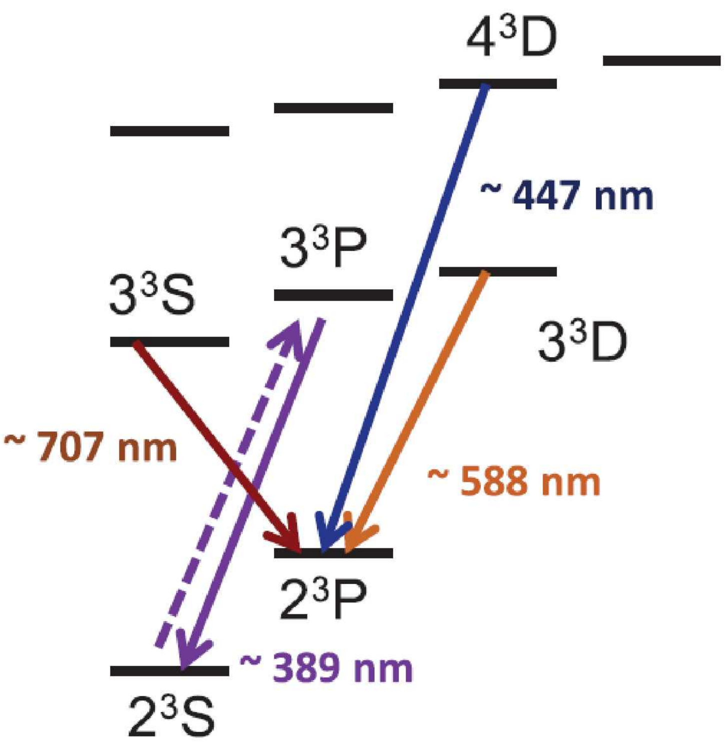
EXPERIMENTAL SETUP





- **Annular powered electrode inside the center tube.**
- **Placing the APPJ in a vacuum chamber - consistent and controlled chemistry, ground planes, and gas flow.**
- **Coaxial tube enables a gas shroud – control environment independently of gas in main jet.**

LASER COLLISIONAL INDUCED FLUORESCENCE (LCIF)

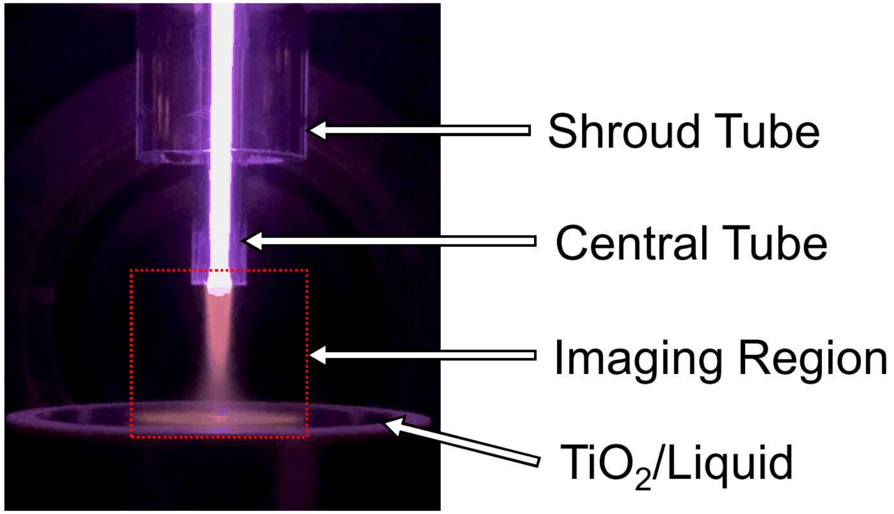


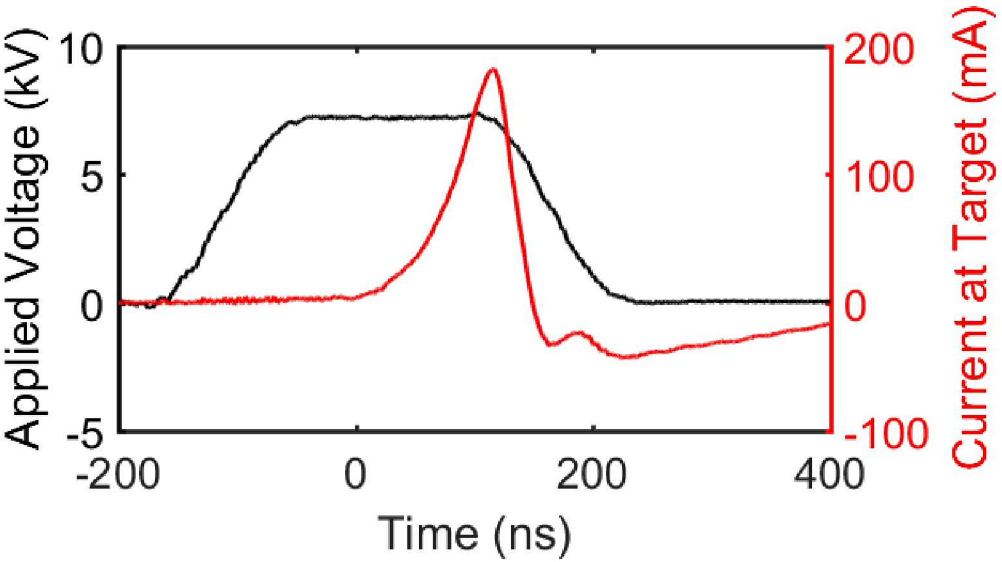
- Barnat and Fierro, J. Phys. D: Appl. Phys., 50, 14LT01 (2017).

- An ultrashort pulse laser (<100 fs) was used to measure the electron density with high time resolution.
- Electrons collide with laser excited He(3^3P)
  - e + He(3^3P) $\rightarrow$ He(3^3D) + e
- n_e is proportional to ratio of LIF signals (588 nm / 389 nm)
- Sufficient He(2^3S) density is critical for accurate LCIF data.
- LCIF was initially developed for pure He, and extended to mixtures for this study of APPJs.

BASE CASE

- +6 kV
- 380 ns pulse, 100 ns rise
- 200 Torr
  - Faster dynamics (for modeling)
  - Lower background LCIF signal (for experiment)
- 500 sccm He in center tube
- 0.75% H_2O in He in shroud, 500 sccm
- Gap to target: 7.5 mm
- Substrate: Liquid water or TiO_2 2.5 mm thick (same $\varepsilon/\varepsilon_0$)
- Current measured at ground electrode under water/TiO_2.





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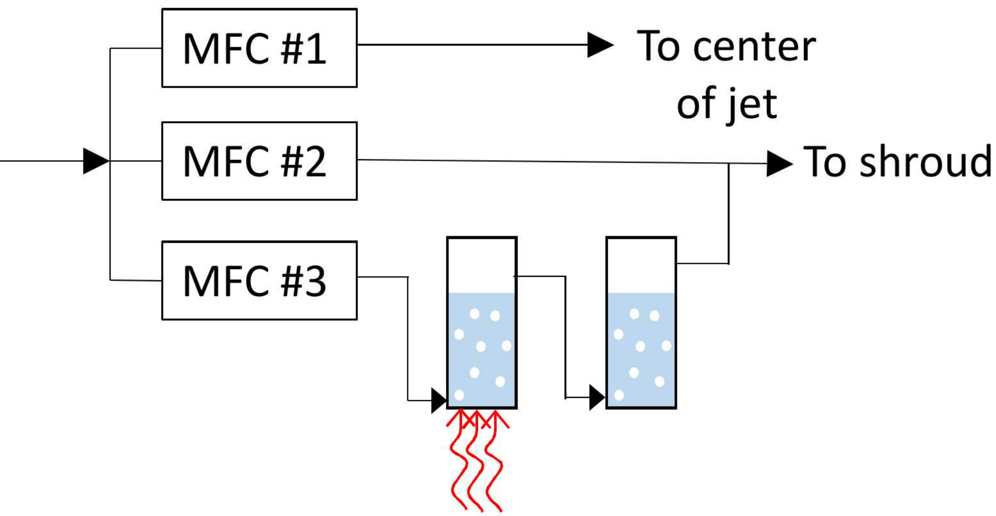
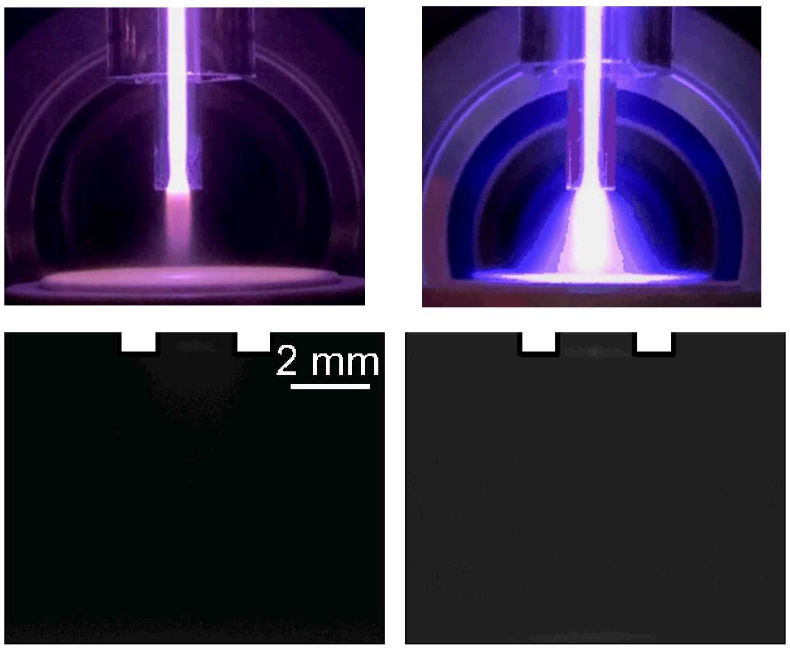
HUMID He SHROUD





- **Humid He shroud confines the jet, much like operating with surrounding air, while being compatible with LCIF measurements.**
- **First bubbler oversaturates water vapor, second bubbler removes excess.**
- **Temperature of second bubbler determines humidity of gas.**

Animation Slide

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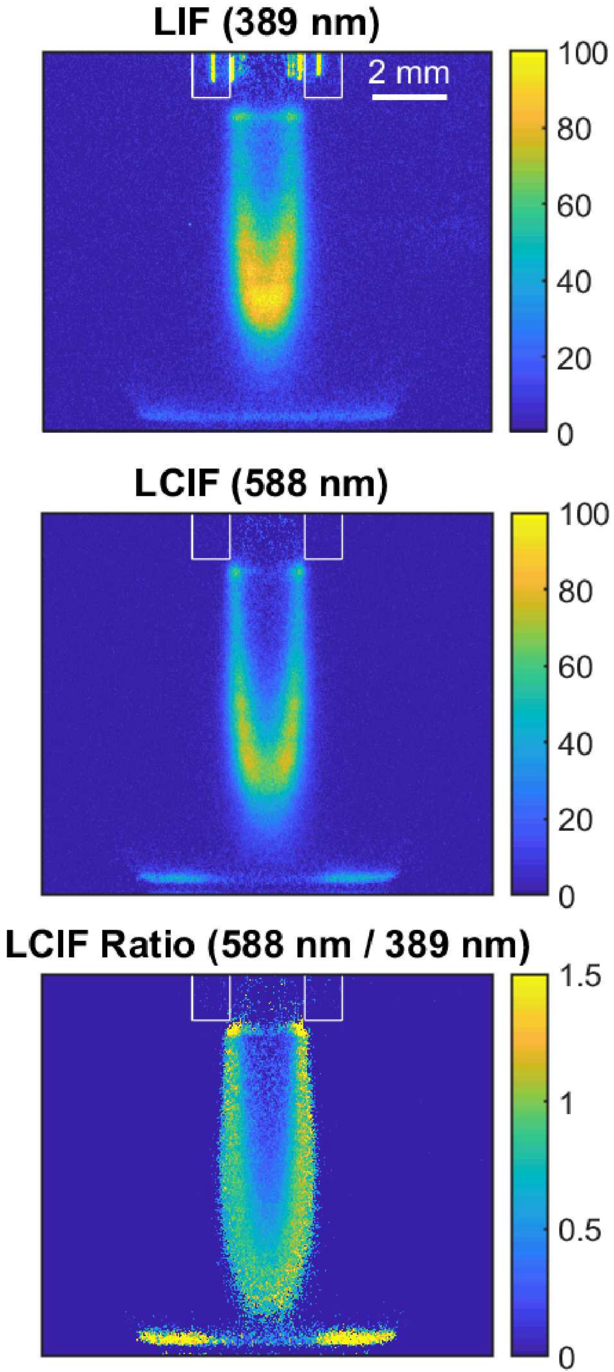

LCIF IN HUMID He

- 6 kV, 430 ns pulse
- Center: 500 sccm He
- Shroud: 500 sccm He/H_2O = 97.7/2.3
- t = 230 ns, 30 ns after IW contacts surface
- Moving away from He core, there are fewer He(2^3S), LIF signal decreases.

$$He(2^3S) + H_2O \rightarrow He + H_2O^+ + e$$

- In regions of high H_2O concentration, there may be significant n_e which is not detectable due to low He(2^3S).

  - Ratio = 1 → $n_e \approx 4\times10^{12}$ cm^{-3}

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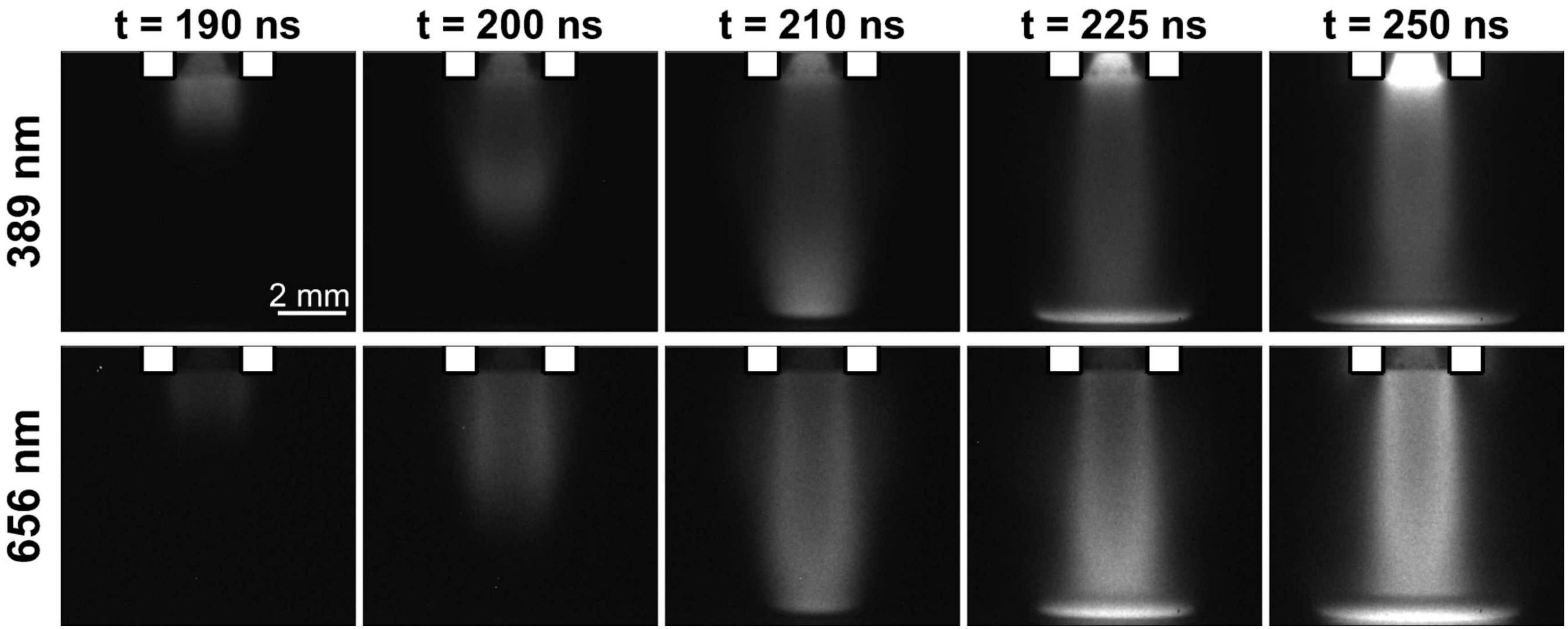
TiO_2: OPTICAL EMISSION



- TiO_2 substrate
- 5 ns ICCD gate, 656 nm – H_α emission, 389 nm – $He(3^3P) \rightarrow He(2^3S)$
- Line of sight imaging (not Abel inverted).
- Even $He(3^3P)$ emission, indicating an annular electron density.
- Surface ionization wave (SIW) forms after IW contacts the surface.

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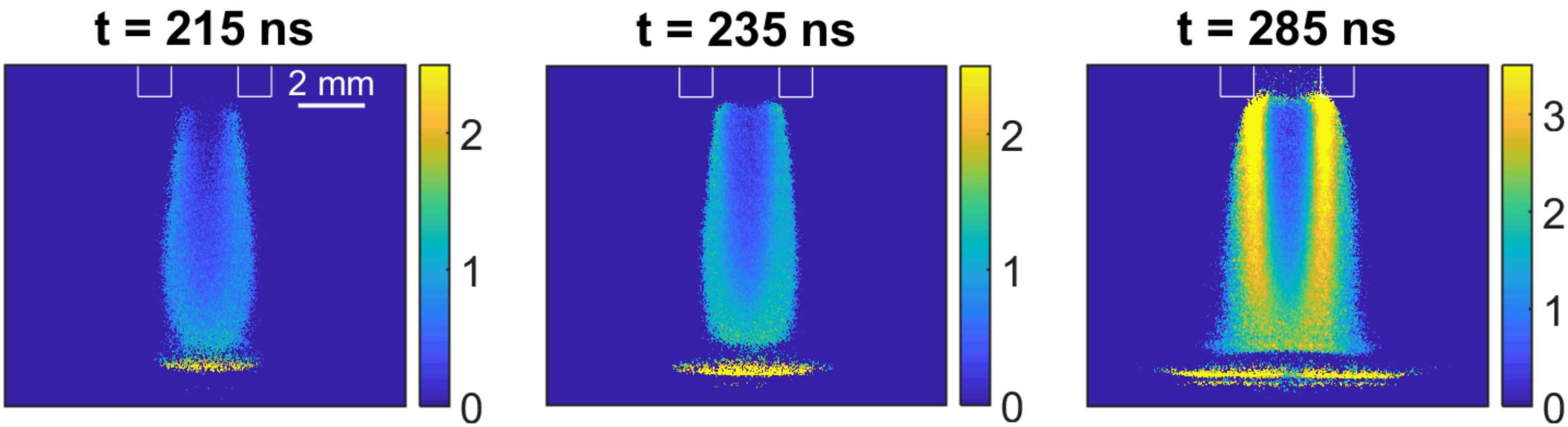
TiO_2: ELECTRON DENSITY



- Electron density profile is annular.
- Photons from the discharge (He^* or He_2^*) ionize H_2O.
- Photoionization selectively occurs in the mixing region of the pure He with the humid He.
- After IW contacts the surface and a restrike occurs, the electron density nearly doubles.
  - Ratio = 1 $\rightarrow n_e \approx 4\times10^{12}$ cm^{-3}

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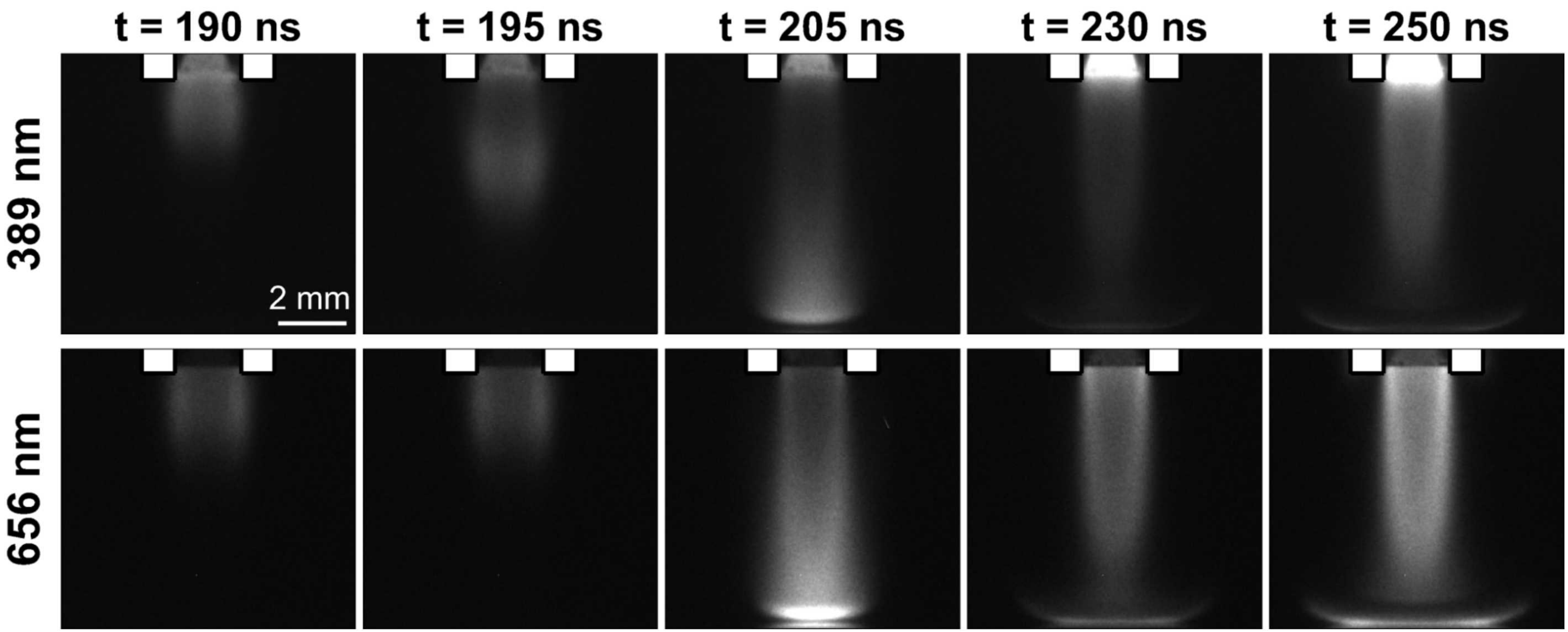
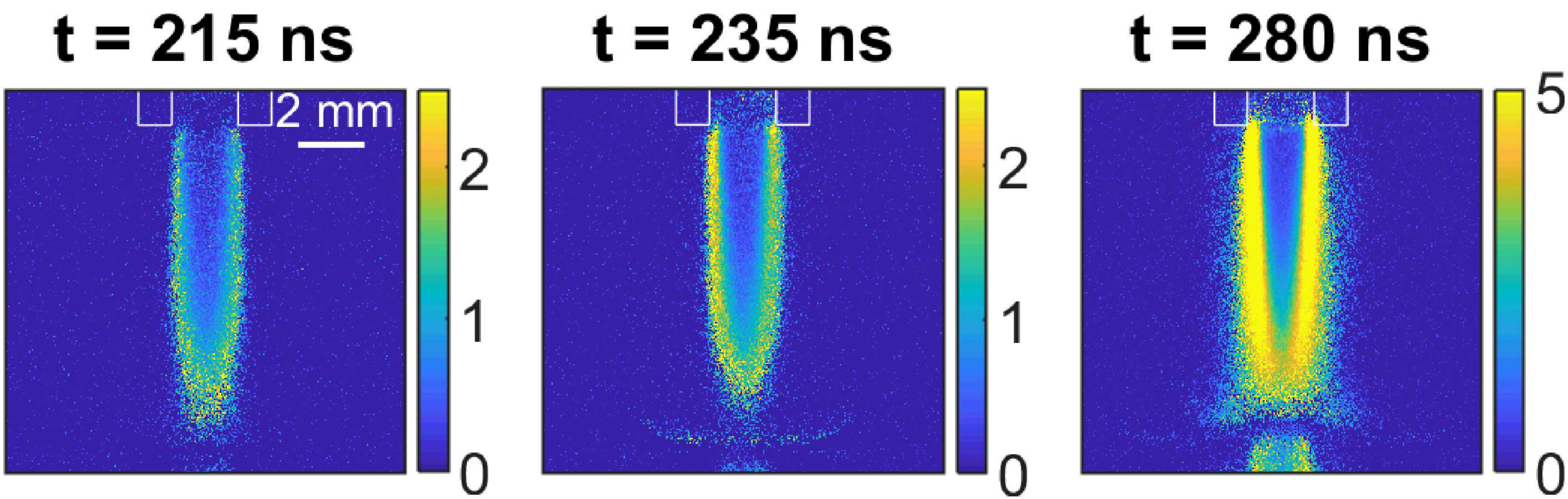
WATER: OPTICAL EMISSION



- Water substrate. 5 ns ICCD gate
- Evaporation results in much higher concentrations of H_2O enabling more photoionization and Penning ionization.
- IW contacts surface 5 ns earlier than for TiO_2 substrate.
- Plasma dims at 230 ns, after contacting the surface.

WATER: ELECTRON DENSITY



- Water substrate.
- Electron density (n_e) is again annular due to photoionization and Penning ionization of surrounding water vapor.
- SIW is not visible by LCIF because high H_2O density near the surface depletes He(2^3S).

$$H_2O + He(2^3S) \rightarrow H_2O^+ + He$$

- Electron density is slightly higher than for TiO_2 because of higher water vapor density.
  - Ratio = 1 $\rightarrow$ $n_e \approx 4\times10^{12}$ cm^{-3}

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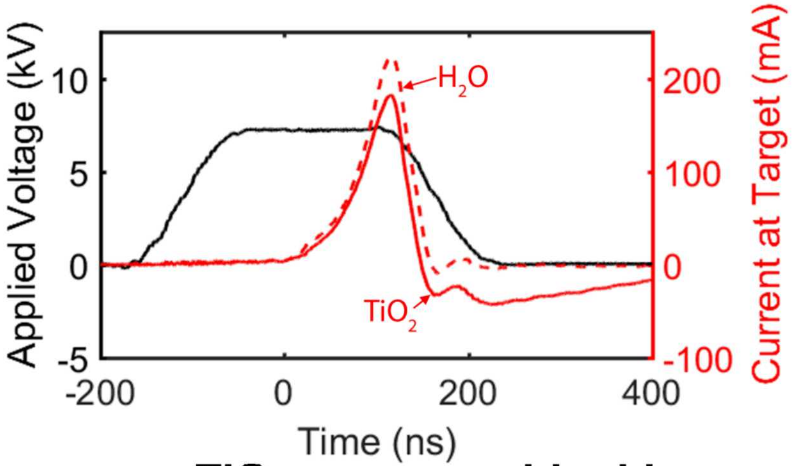
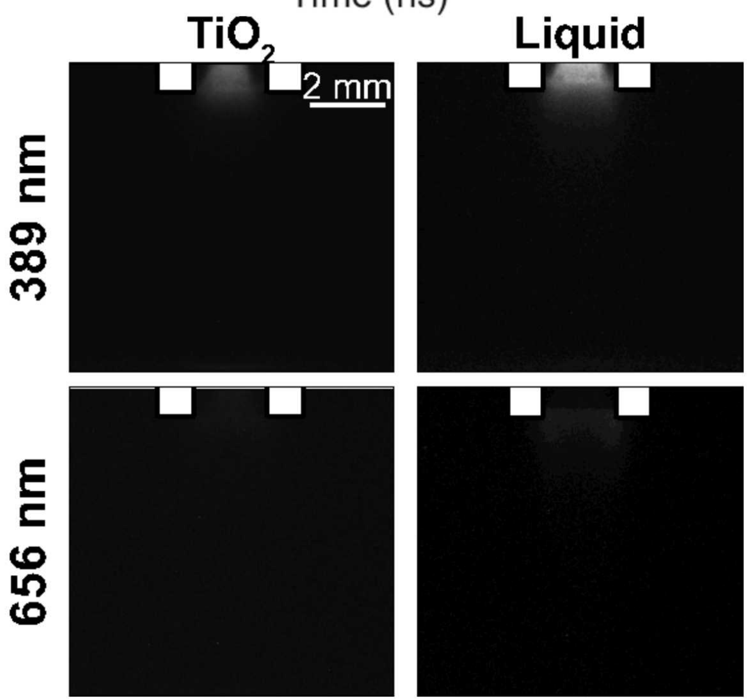
WATER vs. TiO_2

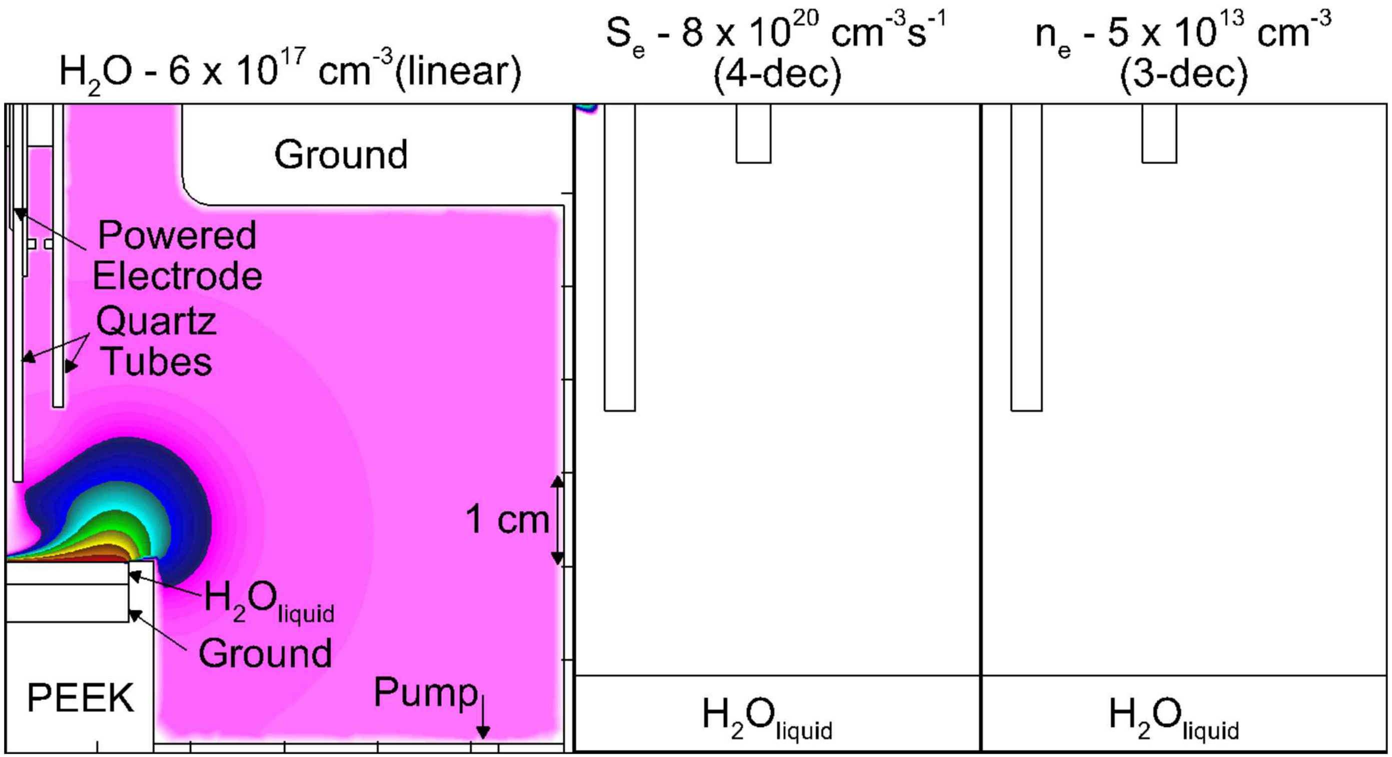




- With water base, magnitude of forward current is higher, reverse current is lower.
- Forward current enhanced by humidity.
- Electrons/ions solvate into the water while being physisorbed on TiO_2.
- Water less likely to support electron emission to support reverse of current.
- Evaporation produces higher water concentrations near surface resulting in lower mobility, thinner SIW.
- Plasma dims after IW contacts liquid surface.

Animation Slide

MODELING LIQUID INTERACTIONS



- **Cylindrically symmetric.**
- **Modeling work ongoing.**
- **Same parameters as experiment.**

- ***nonPDPSIM* – 2-dimensional plasma hydrodynamics model**
- **Photoionization and photoemission from surfaces are critical in positive IW propagation.**
- **Surface ionization wave develops along the liquid surface.**
- **Electron density profile is annular.**

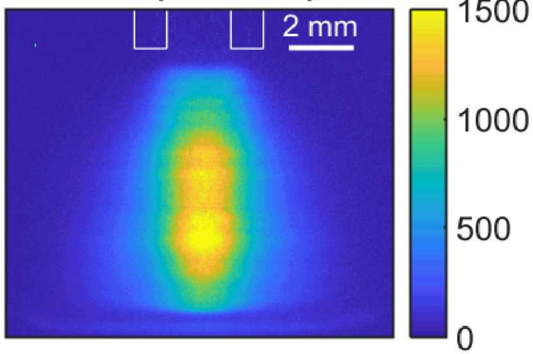
CONCLUDING REMARKS

- An ionization wave exiting a He plasma jet into a humid environment is annular due to Penning ionization and photoionization at the boundary
- Plasma in contact with water vs TiO_2 (same $\varepsilon/\varepsilon_0$) are distinguishable from electrical I-V traces.
  - Mild humidity above the water produces larger forward current.
  - Solvation of electrons/ions in water and larger humidity above water result in lower reverse current.
- SIWs propagating over a liquid surface are thinner due to the lower mobility of electrons in a saturated water vapor environment.

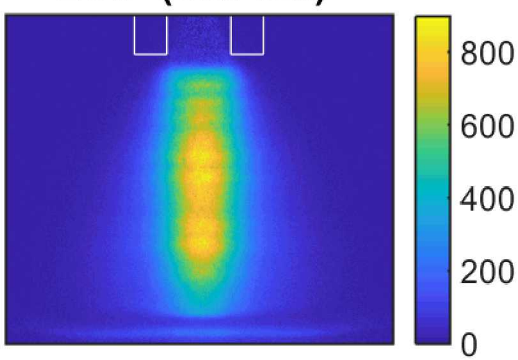
Appendix

CONVERT LCIF TO n_e

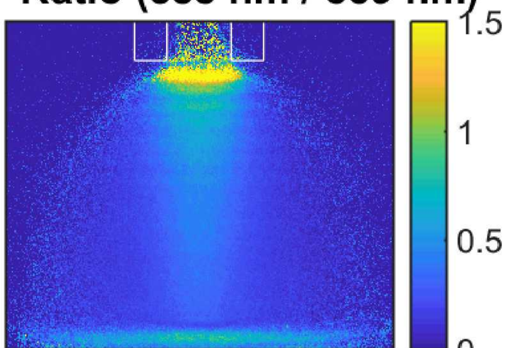
LIF (389 nm)

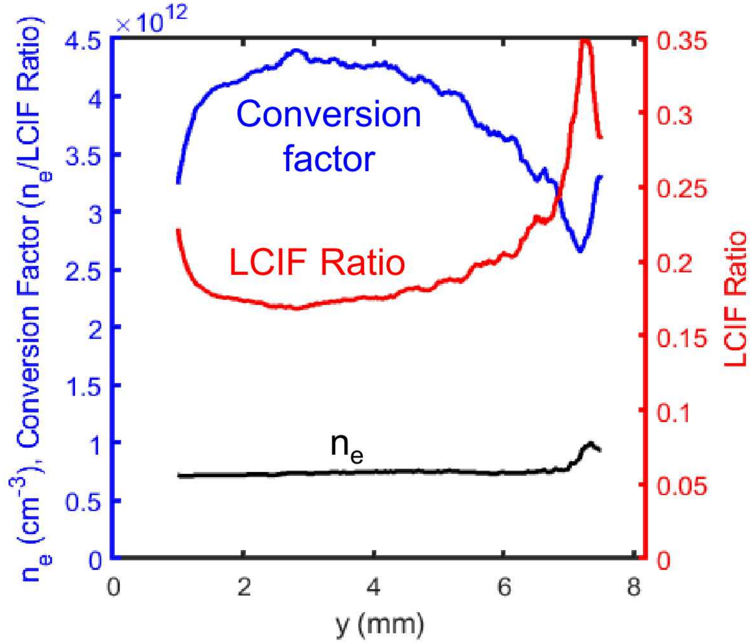


LCIF (588 nm)



LCIF Ratio (588 nm / 389 nm)



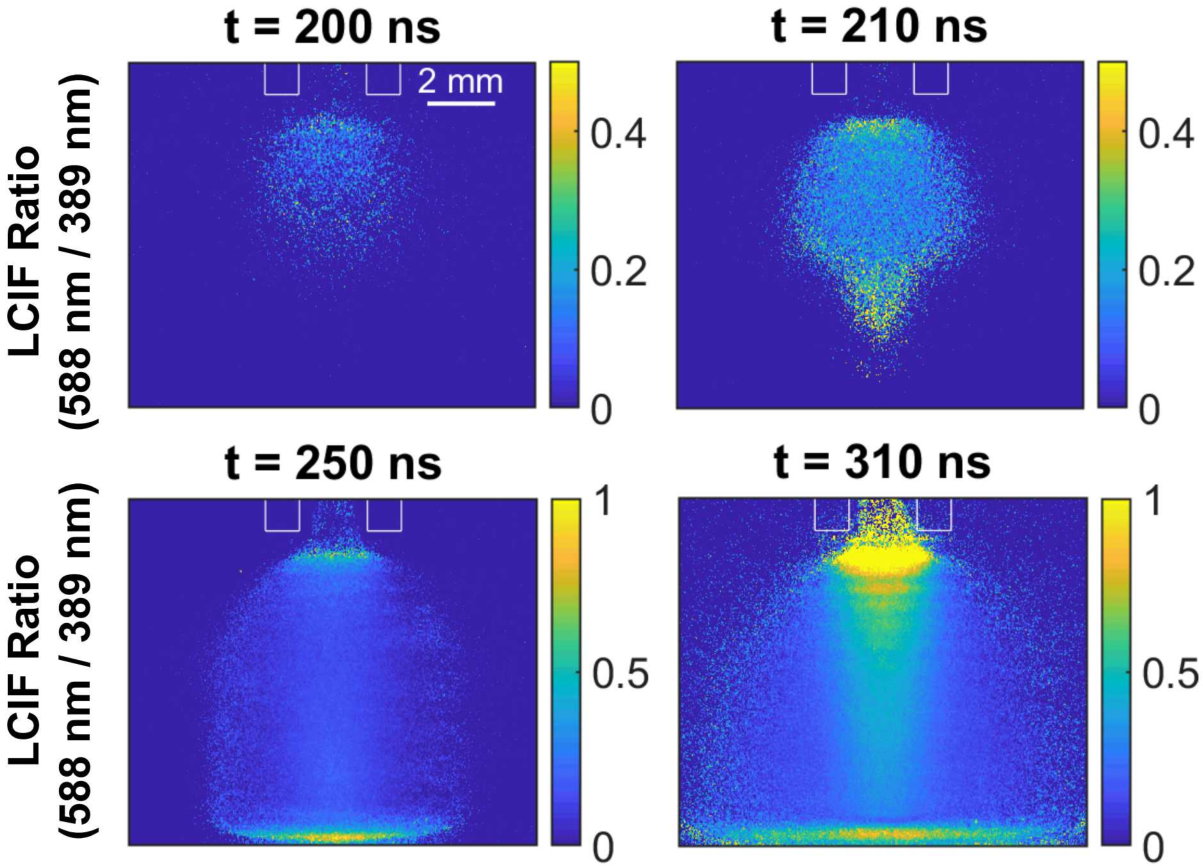


- **Calculate n_e using Ohm's law and E/N.**

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

- **An LCIF ratio of 1, is approximately 4 x 10^{12} cm^{-3} electrons.**
- **Previously, conversion factor estimated at 1.5 x 10^{13} cm^{-3} at 600 Torr.**

BASE CASE LCIF



- +6 kV, 430 ns
- Pure He, 200 Torr

- Before IW reaches the surface, He(2^3S) densities are low.
- n_e in the SIW is nearly double that of the bulk.
- Elevated n_e in IW front may be due to Stark mixing.

Ratio = 1 $\rightarrow n_e \approx 4\times10^{12}$ cm^{-3}

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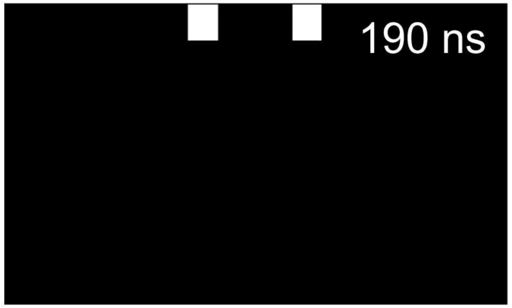
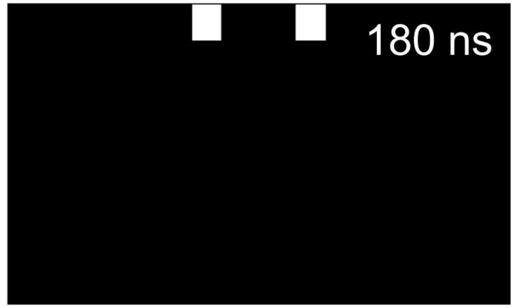
VARY PRESSURE – EMISSION

150 Torr



200 Torr (Base)



300 Torr

400 Torr

500 Torr

600 Torr

- **6 kV, 500 sccm He, 390 nm plasma emission.**
- **350 ns animated, time of first frame indicated.**
- **Varied pulse duration so voltage is on for 80 ns after contact.**
- **IW propagates slower and SIW becomes thinner for higher pressures. (lower electron mobility)**

Animation Slide

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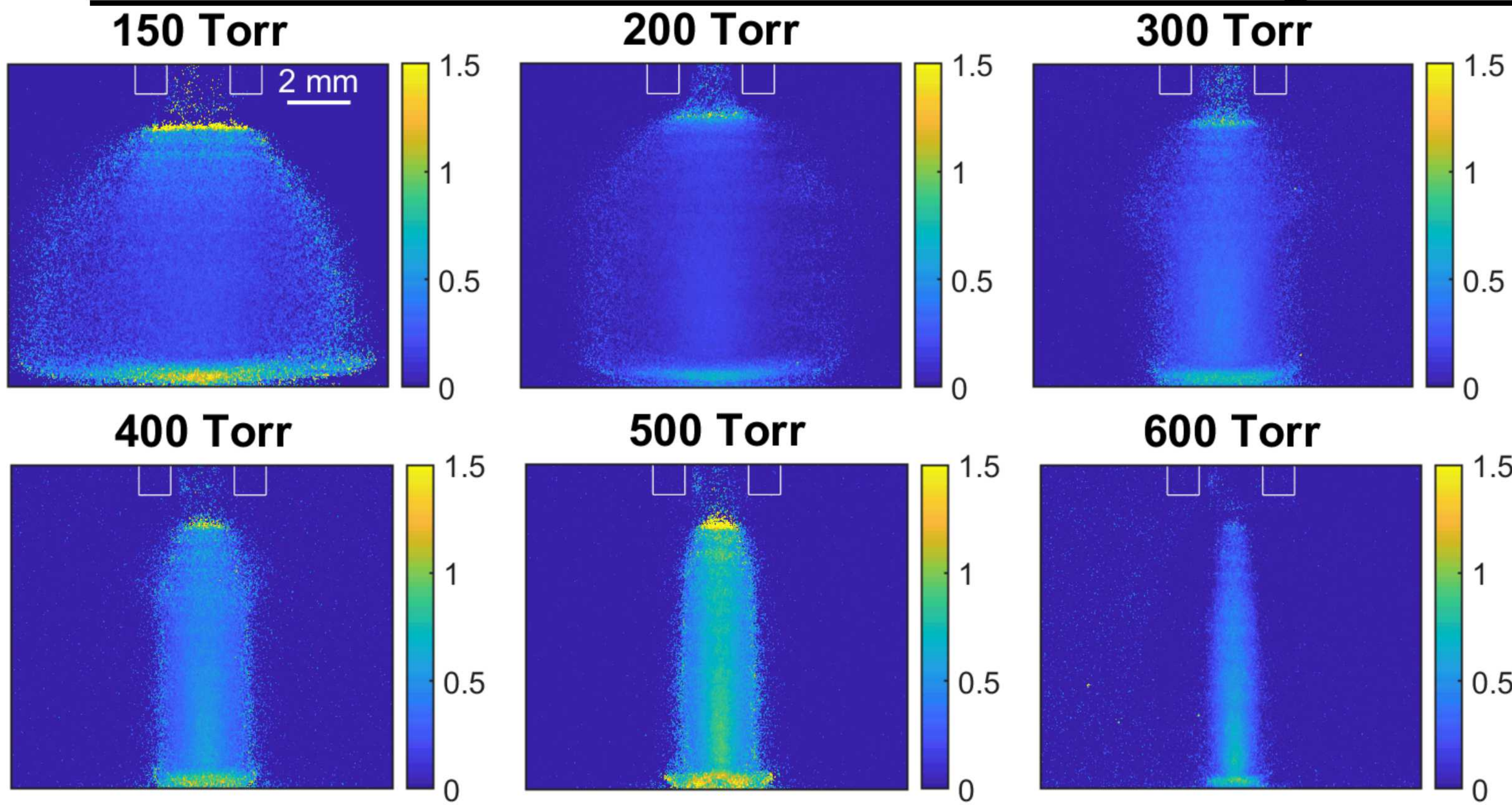
VARY PRESSURE – n_e



- 6 kV, 500 sccm He, 30 ns after IW contacts surface.
- Plasma is more confined at higher pressure, n_e increases.
- Current and energy deposition decrease with increasing pressure.
- Above 500 Torr, n_e is collisional enough that ionization rate drops.

Ratio = 1 $\rightarrow n_e \approx 4\times10^{12}$ cm^{-3}

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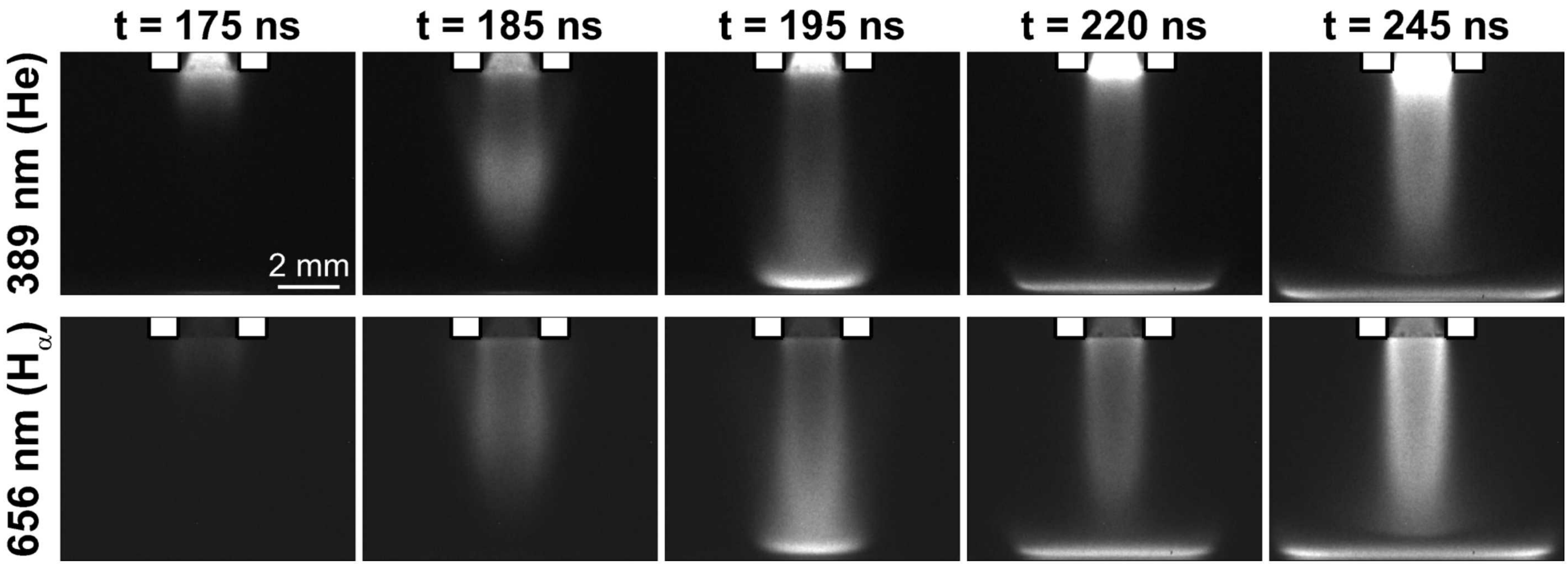
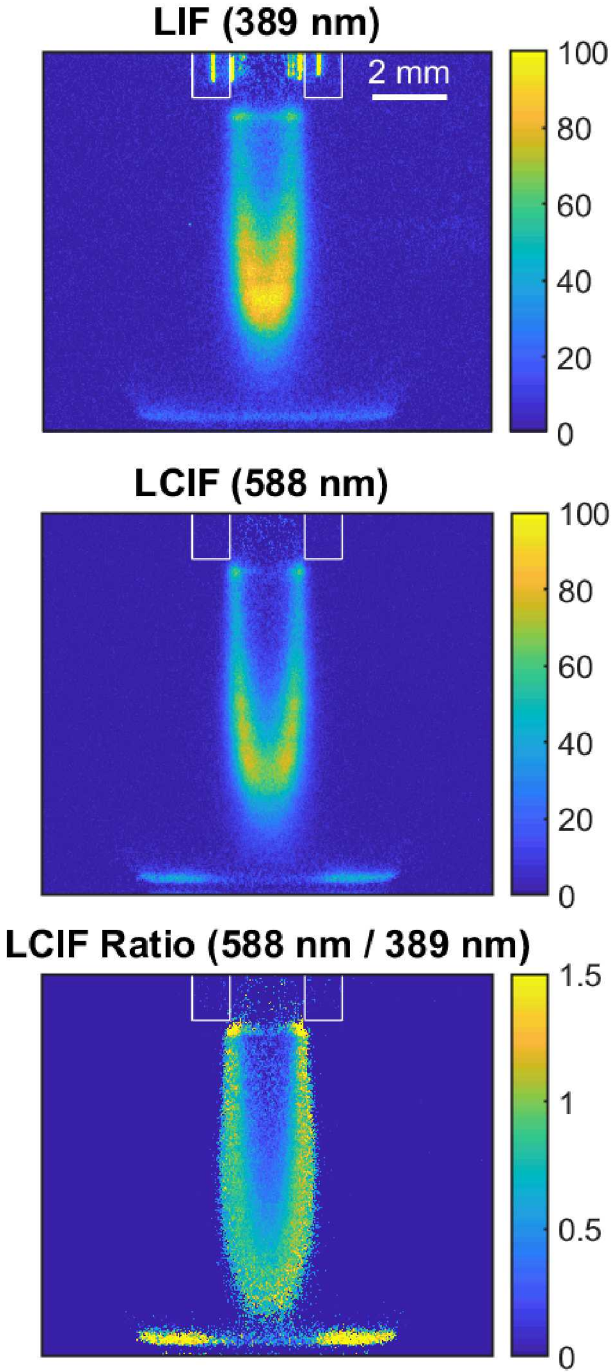
HUMID He SHROUD EMISSION



- 2.3% H_2O in shroud.
- Images have not been Abel inverted.
- IW reaches outlet of the tube earlier than in base case – photoionization from He_2^* causes non-local seed ionization.
- Photoionization and Penning ionization promote IW speed.
- H_α emission appears more annular – dominates at the interface of the center and shroud flow.

Animation Slide



HUMID He SHROUD

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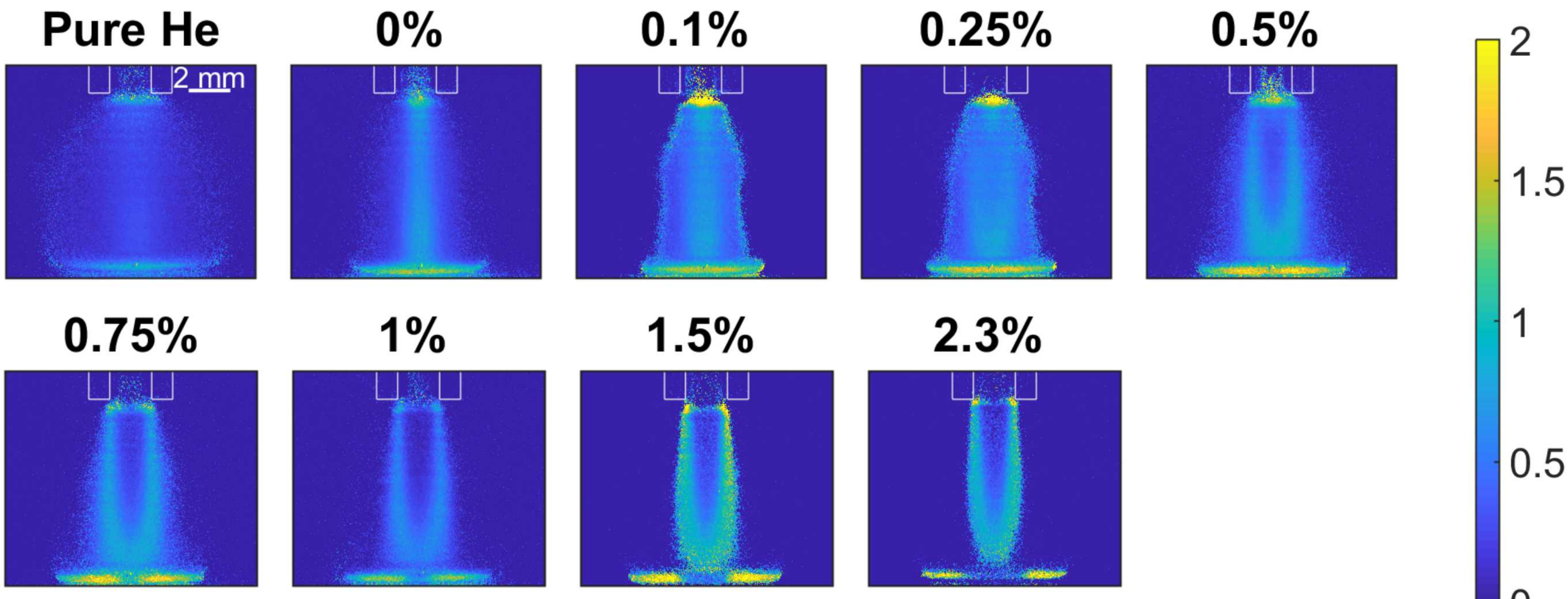
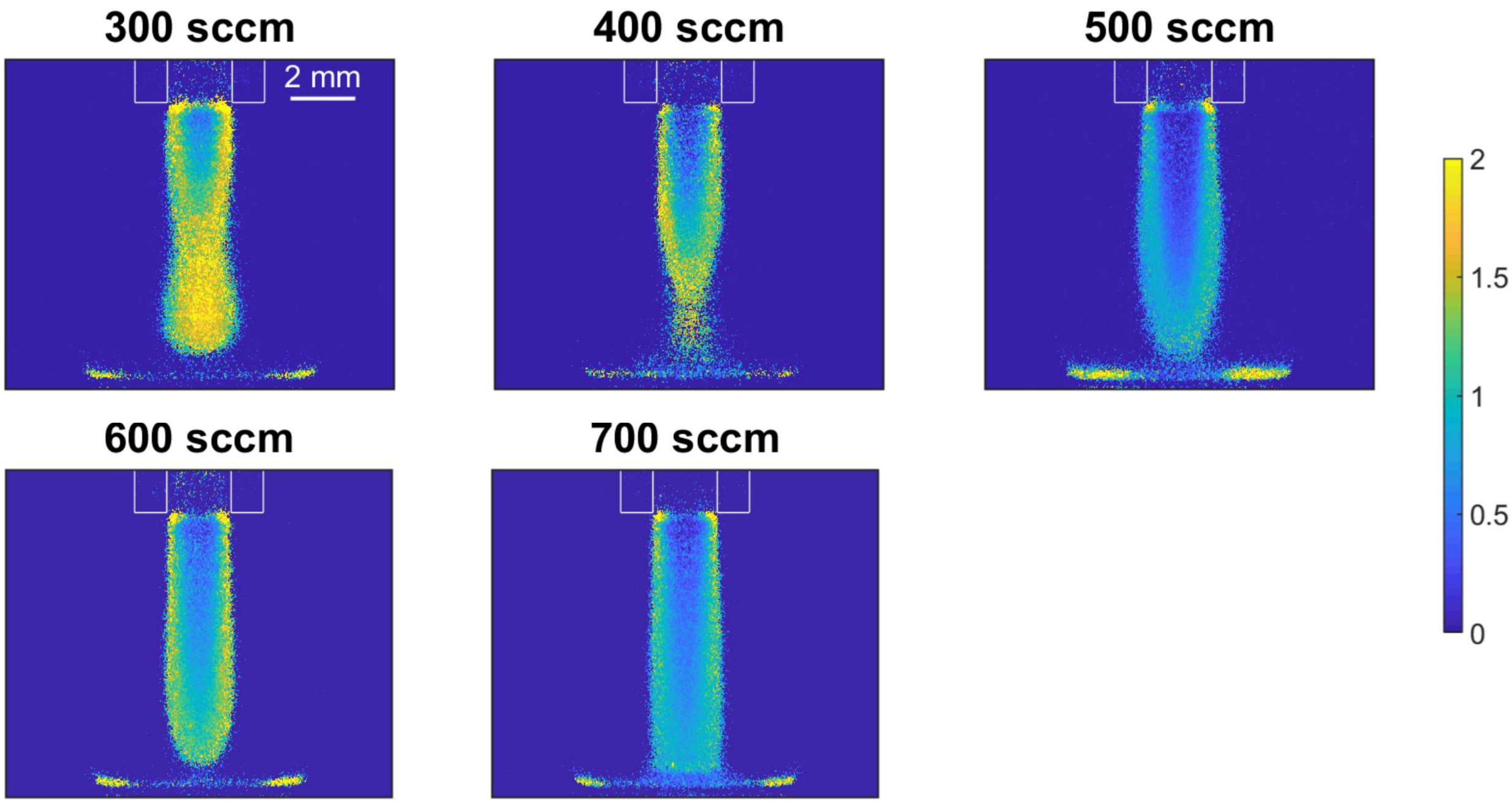
SHROUD HUMIDITY



- 30 ns after IW reaches surface.
- Transition from diffuse in pure helium case to confined by humid shroud.
- Higher electron energy loss rates with H_2O because of vibrational and rotational excitation.
- n_e increases with humidity due to Penning ionization.

Ratio = 1 $\rightarrow n_e \approx 4\times10^{12}$ cm^{-3}

VARY He FLOW RATE - n_e



- Vary center flow rate.
- Shroud: 500 sccm, He/H_2O = 98.7/2.3
- Higher He flow rates more rapidly convect in-diffusing H_2O.

Ratio = 1 $\rightarrow n_e \approx 4\times10^{12}$ cm^{-3}

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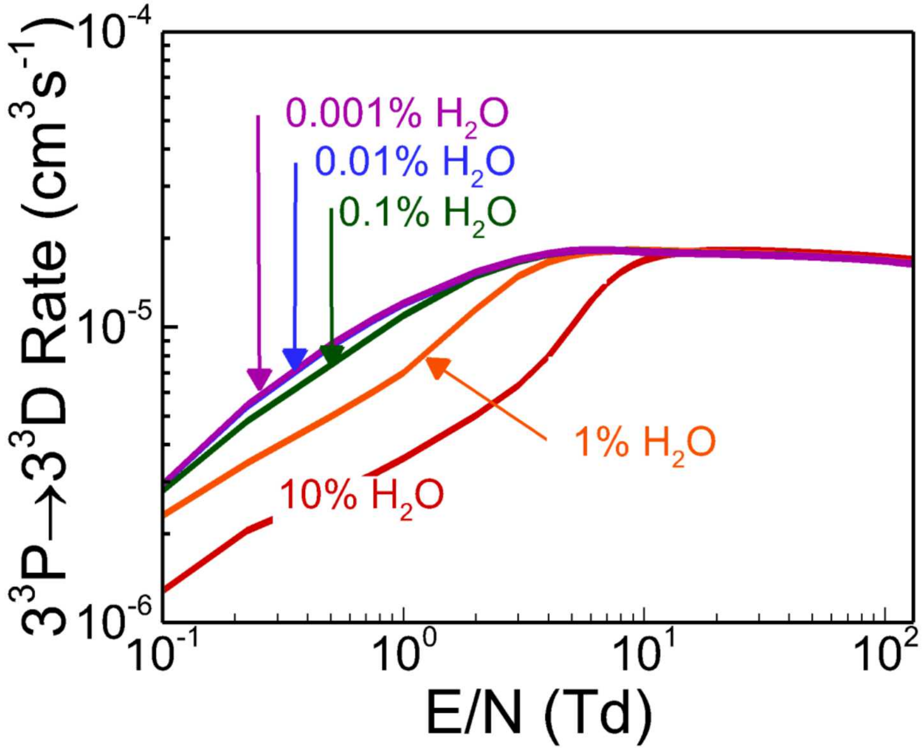
BOLTZMANN CALCULATIONS



- **For LCIF measurement to be a linear representation of n_e, this rate must be independent of T_e:**

e + He(3^3P) $\rightarrow$ He(3^3D) + e

- **Threshold = 0.06 eV**
- **In pure He, this occurs when E/N > ~ 0.8 Td**
- **For H_2O < 1%, LCIF is valid ~ 1 Td**