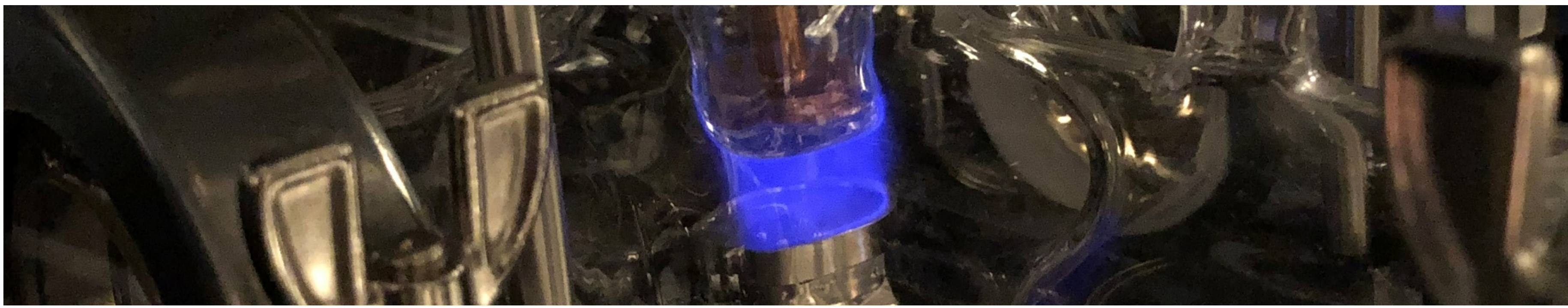
SAND2021-10906C

# 2D-IMAGING OF METHYL IN AN $N_2/CH_4$ NANOSECOND PULSED PLASMA BY PHOTO-FRAGMENTATION LASER INDUCED FLUORESCENCE



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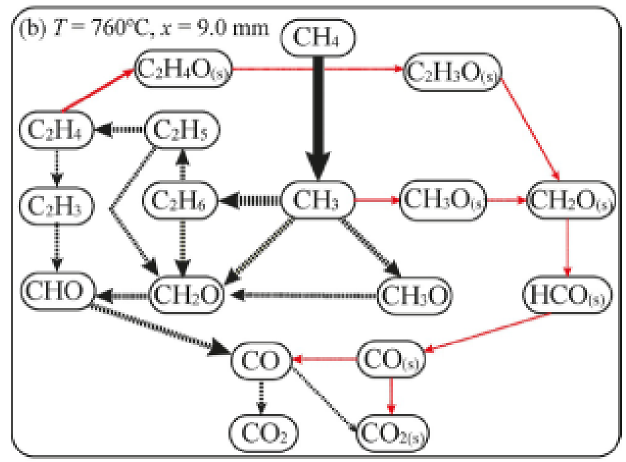
# Methyl as entry point for Methane coupling reaction kinetics

- Methyl important radical in ***methane coupling*** (e.g. **O**xidative **C**oupling of **M**ethane, **OCM**)

- Difficult due to ***methane stability*** → Plasma promising for hydrogen abstraction

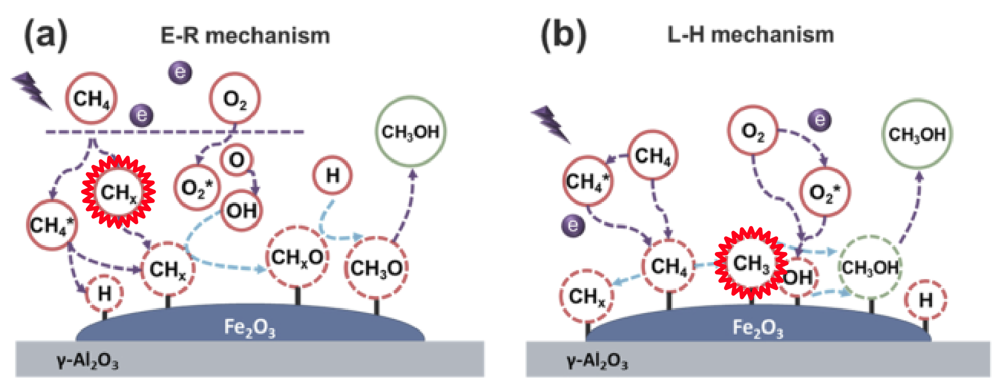
- Importance of **gas** vs. **surface** reactions (catalysts)?

- 2D-Imaging of methyl important but challenging…




red = surface reaction
black = gas phase reaction

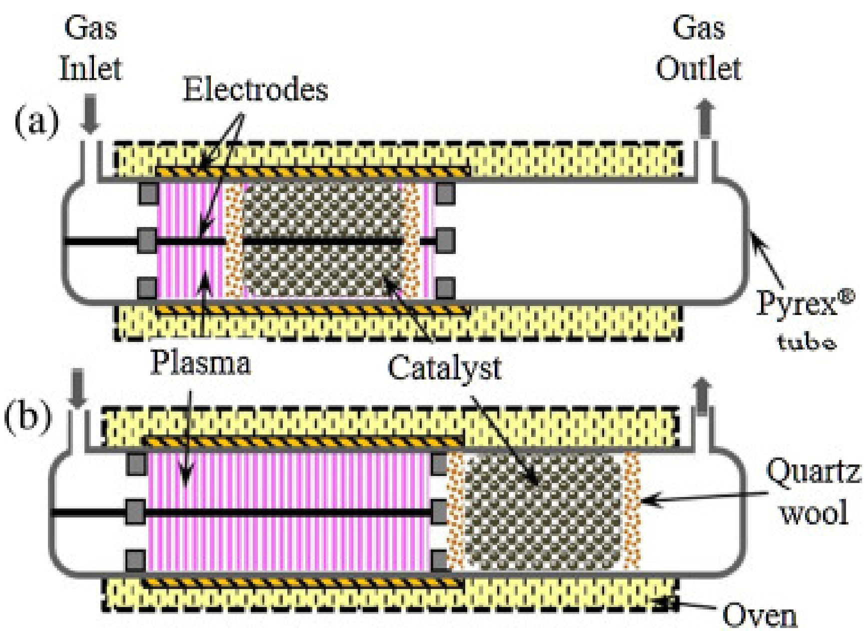
Karakaya & Kee, ChemCatChem **9**, 4538 (2017)



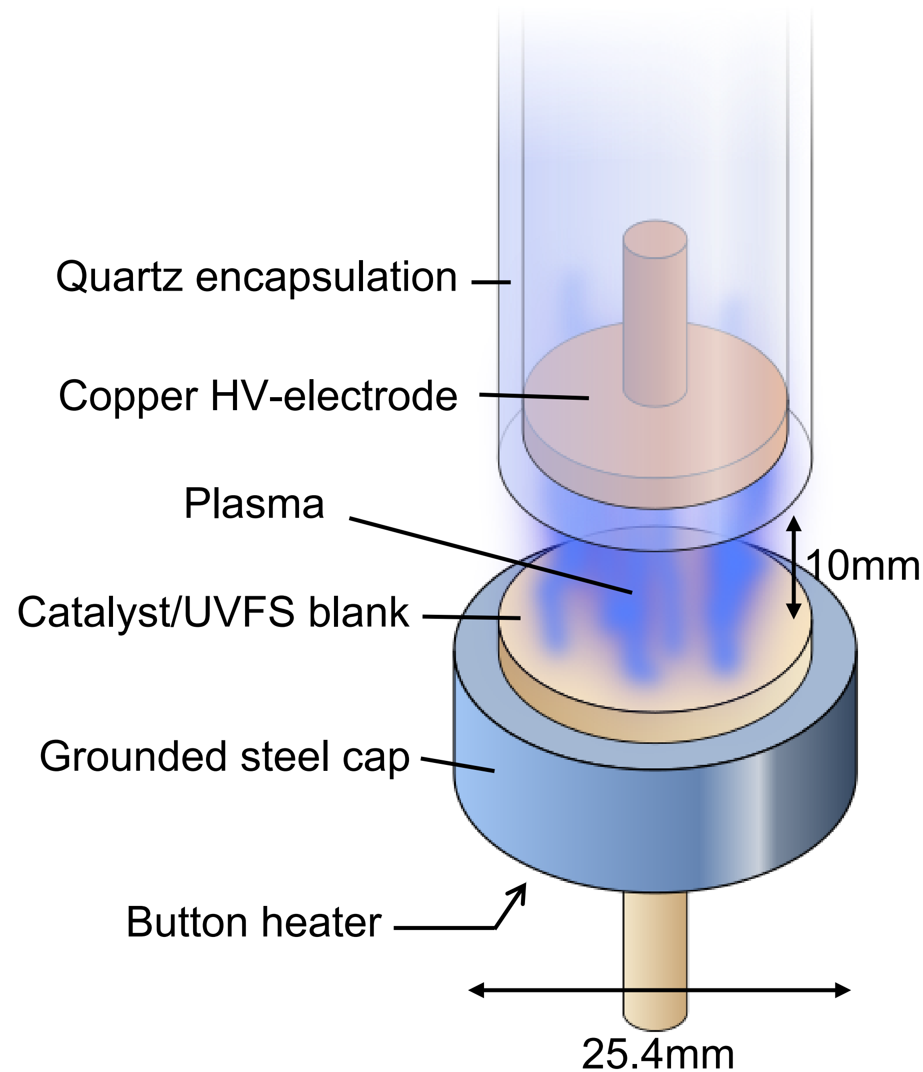

P. Chawdhurry et al., Appl. Catal. B: Env. **284**, 119735 (2021)

# Plasma Configuration

- Plasma + catalyst coupling: Tradeoff Reactivity vs. diagnostic access
- *Our design*: Direct plasma + catalyst interaction with good optical access




T. Pham Huu et al. Catal. Today **257**, 86 (2015)

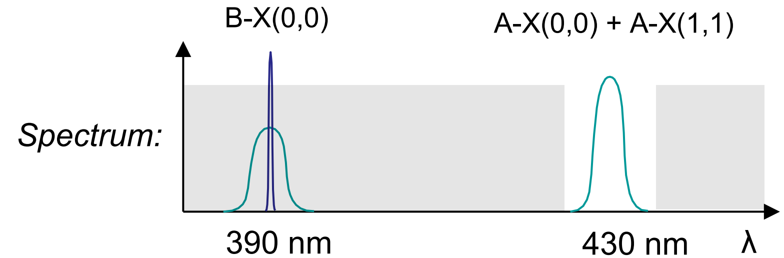


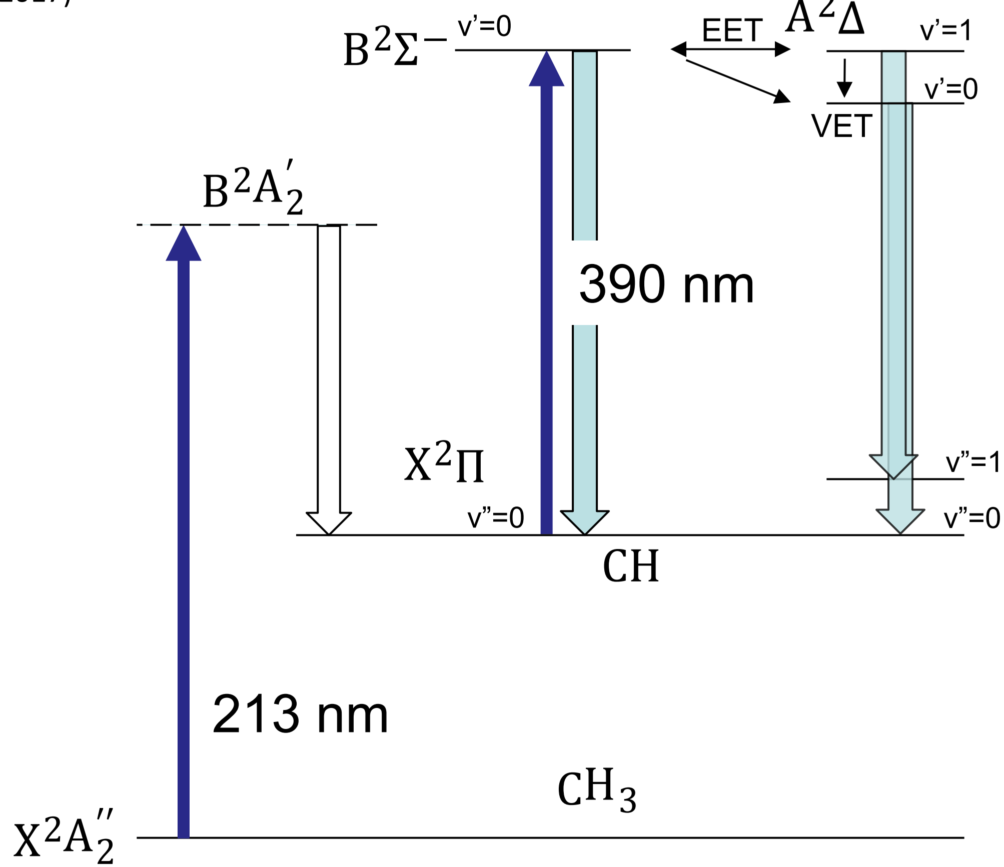
# Photo-Fragmentation Laser Induced Fluorescence

- $CH_3$ difficult to detect with LIF directly → ***Photo-fragmentation LIF***

Li et al. Proc. Combust. Inst. **37**, 4487 (2017)

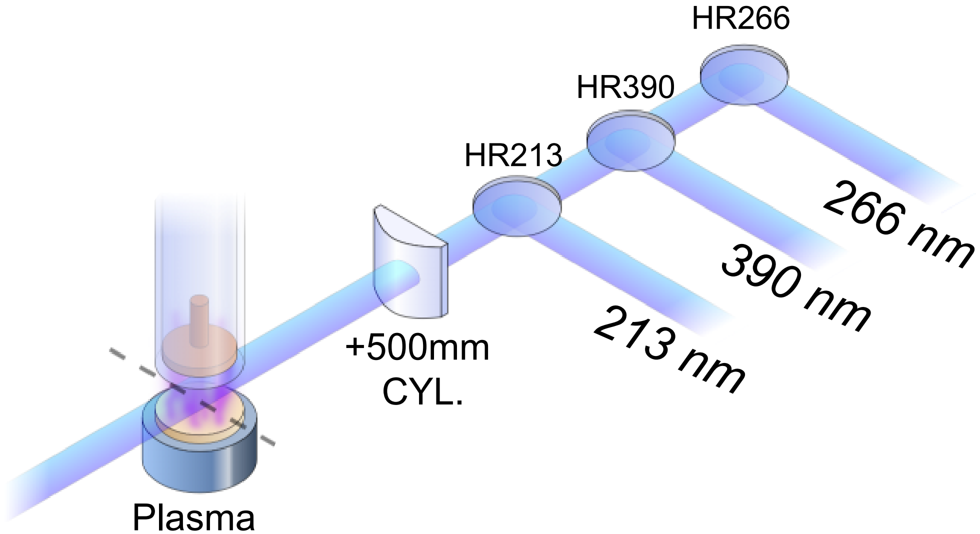
- Detection close to surface → need ***non-resonant detection***
- Excite **B**-state and rely on Electronic Energy Transfer (EET) to populate **A**-state
- EET is collisional quenching process, so collision partner dependent!



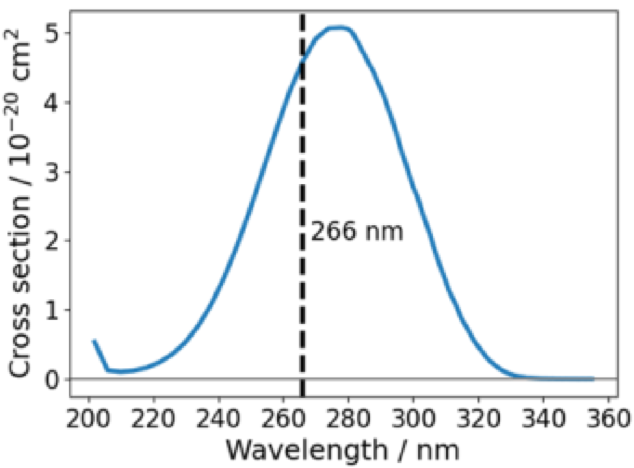



# Optical Layout

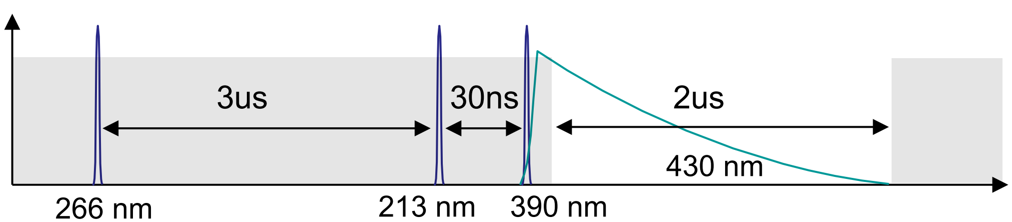
- PF-LIF lasers:
  - **PF:** 213 nm (5th HG YAG)
  - **LIF:** 390 nm (1064nm + 616nm dye)
- Methyl source → **P**hoto-**D**issociation of acetone:
  - **PD**: 266 nm (4th HG YAG)



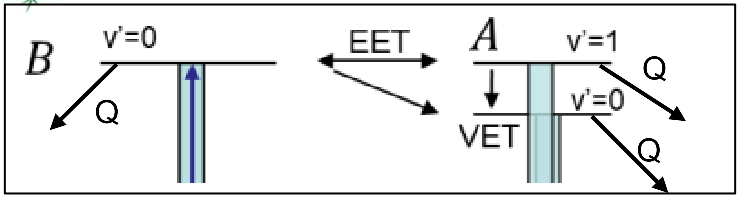

*Acetone absorption:*



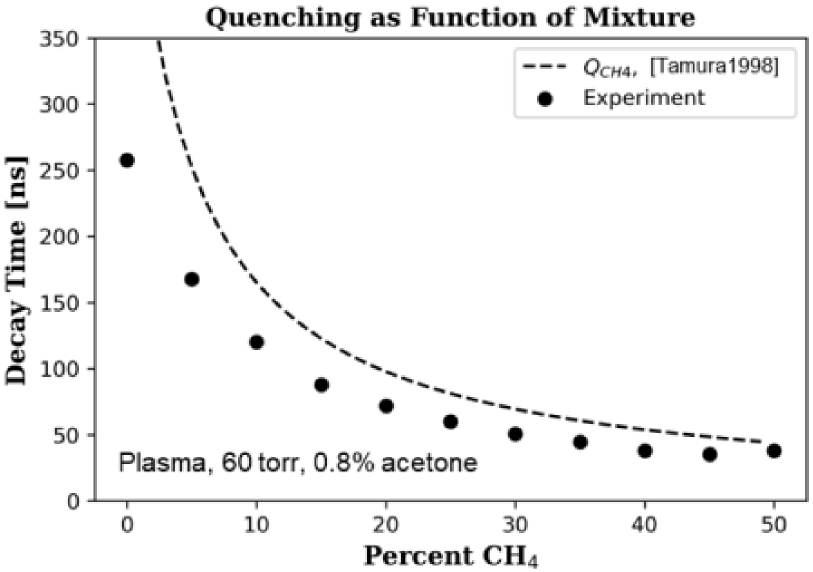

- PMT & spectrometer for characterization
- ICCD + Bandpass filter for imaging
  (430 nm CWL, 10 nm FWHM)



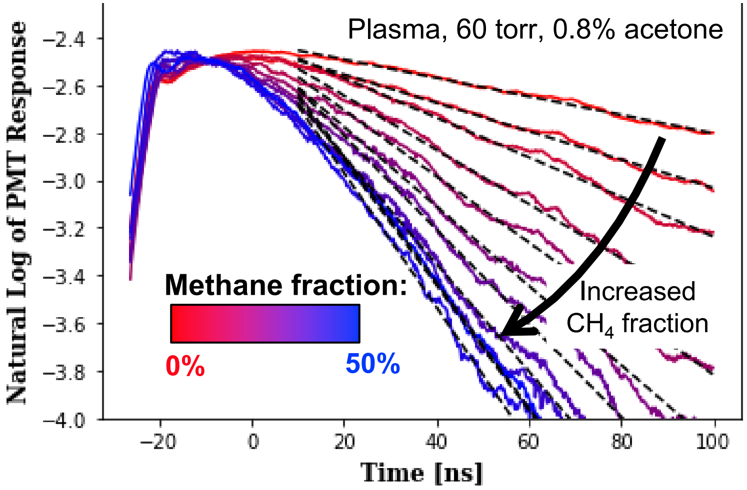
# Calibration: Quenching & yield




- Quenching of 3-level system very complex → Perform **calibration** & **experiment** under ***identical quenching conditions***
- ***Approach***: Increase methane fraction until it dominates quenching




M. Tamura et al. Comb. & Flame **114**, 502-514 (1998)




- At 50% $CH_4$, decay time is not affected by acetone or plasma:

  → ***Quenching dominated by $CH_4$!!***
- For CH-quenching, temperature dependence divides out

# Calibration: Temperature correction

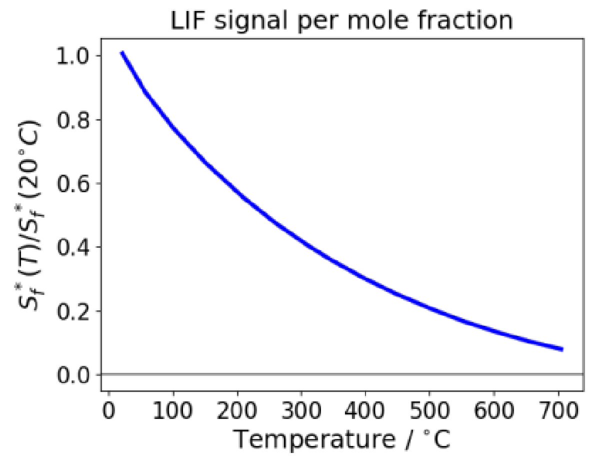
- Photo-dissociation of acetone:

$$(CH_3)_2CO + h\nu \rightarrow CH_3 + CH_3CO$$

- Methyl density given by:

Methyl density · Acetone density · Laser fluence · Absorption cross-section · Photon energy · Methyl yield

$$n_{CH_3}(\vec{r}) = n_{ace}(T(\vec{r}))\,\frac{F_{266}(\vec{r})\,\sigma_{abs}(T(\vec{r}))}{h\nu}\,Y_{CH_3}$$

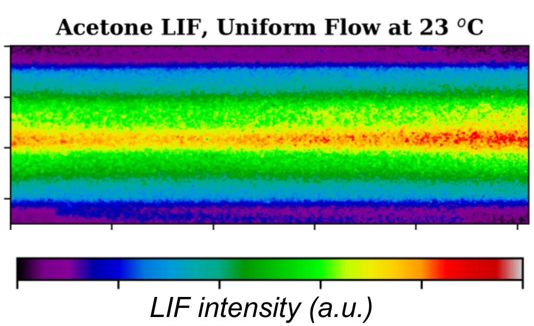



M. Thurber et al. Appl. Optics **37**, 21 (1998)

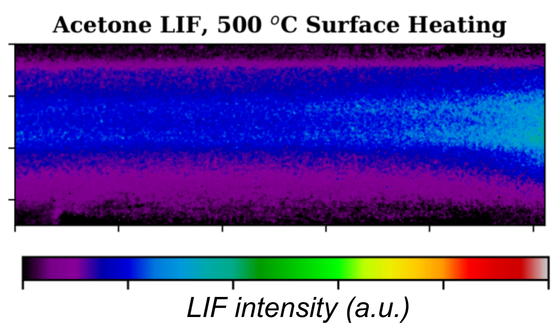
...Temperature profile $T(\vec{r})$ needed!

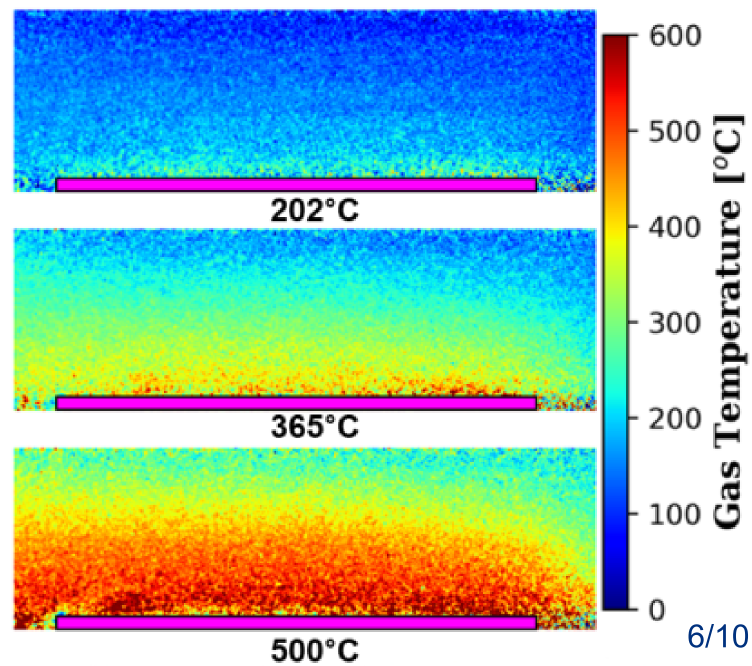
- Acetone LIF intensity function of temperature
  → Determine $T(\vec{r})$ from ratio
  $I_{LIF}(\vec{r}, T)/I_{LIF}(\vec{r}, 20°C)$

$I_{LIF}(\vec{r}, 20°C)$




$I_{LIF}(\vec{r}, T)$



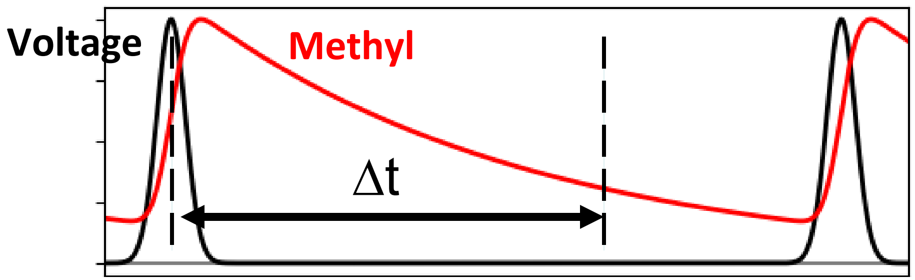



# Absolute methyl profiles

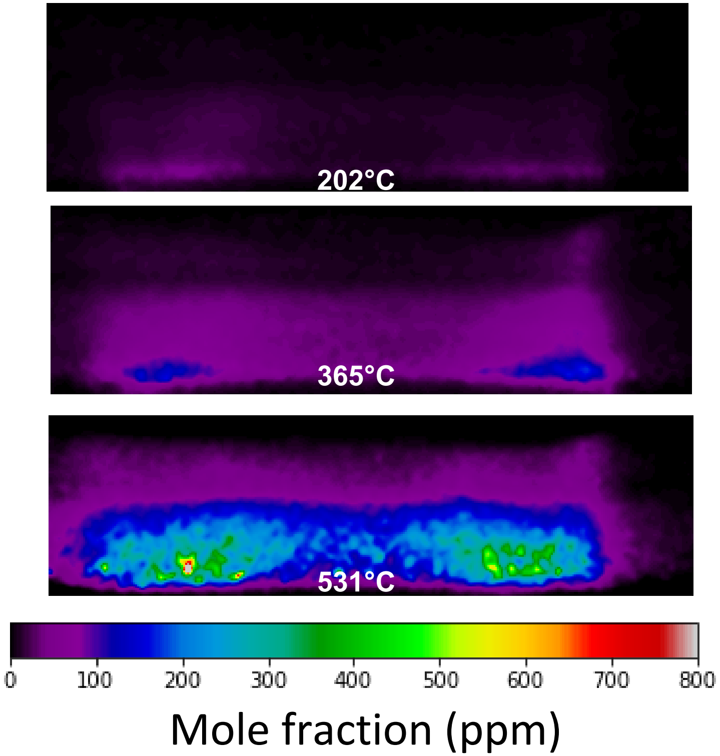
Plasma conditions:

- 20kV, 90ns pulse width @ 200 Hz
- 60 torr, 2 slm
- 50% $N_2$, 50% $CH_4$ (no acetone)

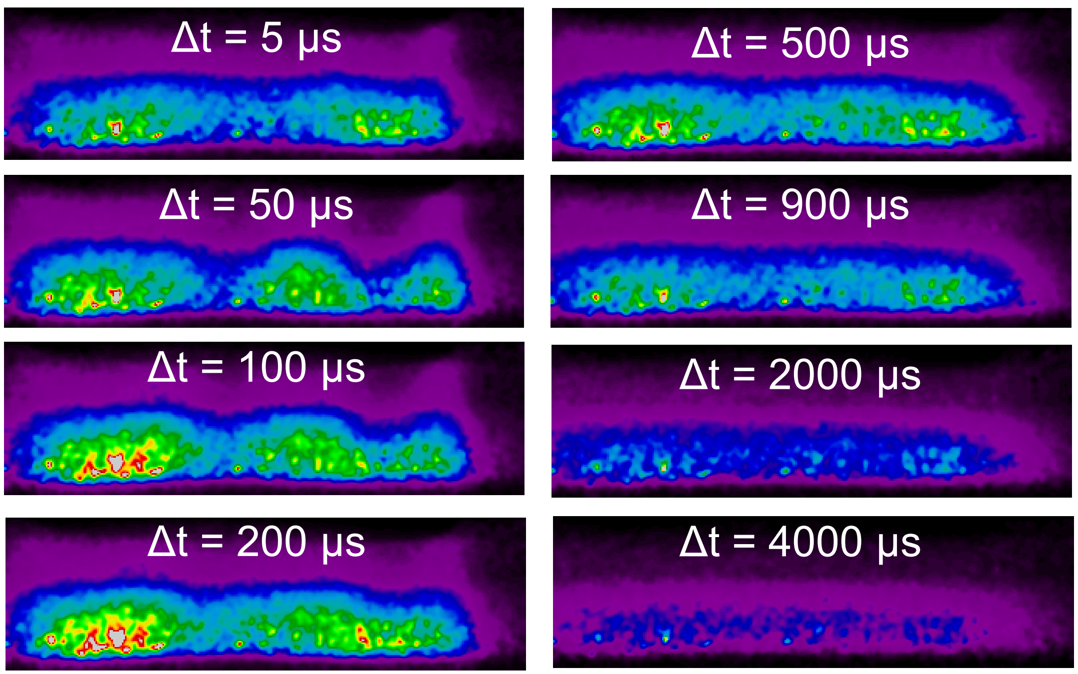
*Temporal scan:*




*Temperature scan:*




*T = 531°C*



# Conclusions

- Photo-Fragmentation Laser Induced Fluorescence was demonstrated for detection of 2D-methyl profiles in a plasma
- Non-resonant LIF scheme allows detection near a surface
- Methyl produced by photo-dissociation of acetone was used for absolute calibration
- Quenching dominated by methane doesn't vary between calibration and experiment
- Acetone LIF was used for temperature corrections
- Absolutely calibrated time resolved 2D-methyl profiles were measured in a ns-plasma, obtaining methyl concentrations up to 800 ppm

# Acknowledgements

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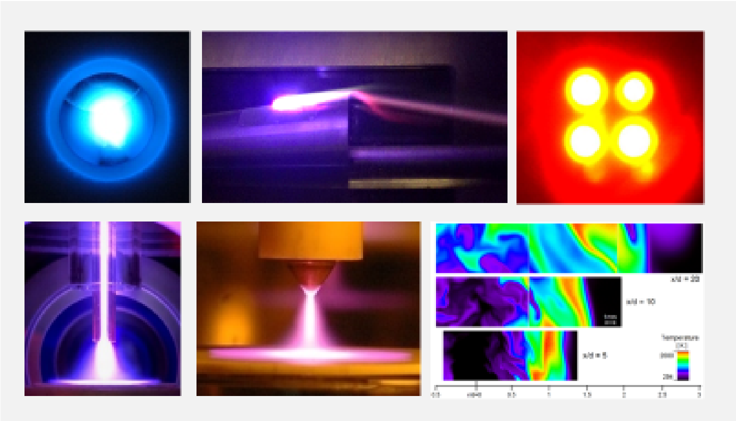
# Thank you!

**This work is part of the**
***Low-Temperature Plasma Research Facility***

The Sandia Low-Temperature Plasma Research Facility (PRF) offers collaborators access to cutting edge diagnostic and computational capabilities and the expertise that is needed to set up and execute experiments and analyze data generated during the collaborative endeavor



**…Interested in collaborating? → Contact us!**

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**Next call: December 2021**