Spatial Structure and Expansion Dynamics of Laser-produced Cerium Plasmas

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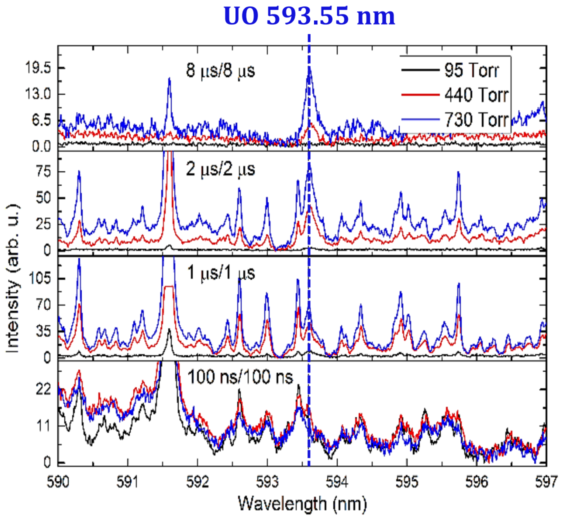
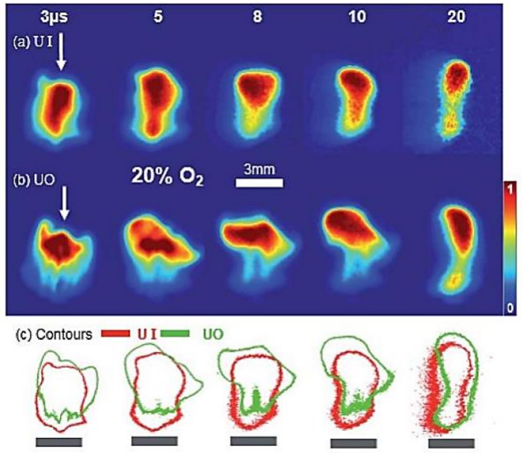
Introduction and Motivation

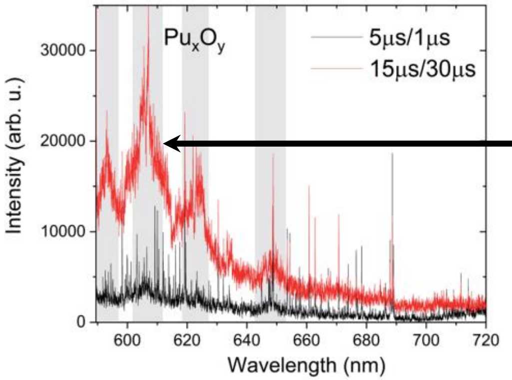
- Optical techniques have repeatedly been used to
  - Detect uranium in the environment
  - Study the physical and chemical properties of uranium nuclear fireballs using laser-produced plasmas (LPPs) surrogates
- Plasmas produced by laser ablation (LA) are
  - Characterized by initial temperatures of ~2 eV
  - Highly transient and sensitive to their surrounding environment
  - Molecular species formation is greatly influenced by that environment and can considerably impact particle formation and debris distribution following the detonation of a nuclear device
- Literature is very limited regarding laser-produced plasma surrogates of plutonium nuclear fireballs



K.C. Hartig *et al., Sci. Rep.* **7** (2016).



E.J. Kautz *et al., J. Anal. At. Spectrom.* **34**, 2236 (2019).

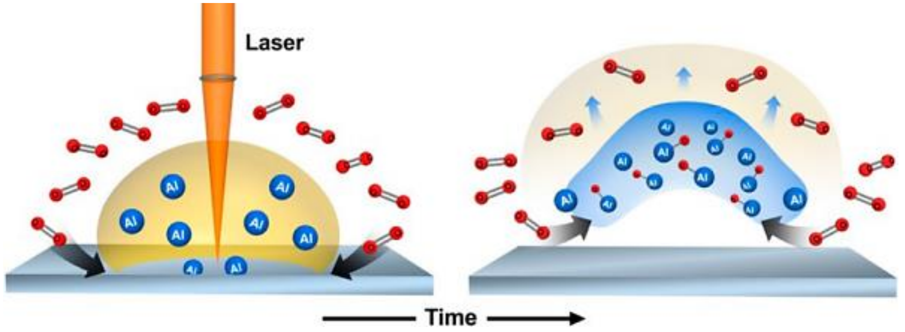


Identification of Pu_xO_y bands is inconclusive

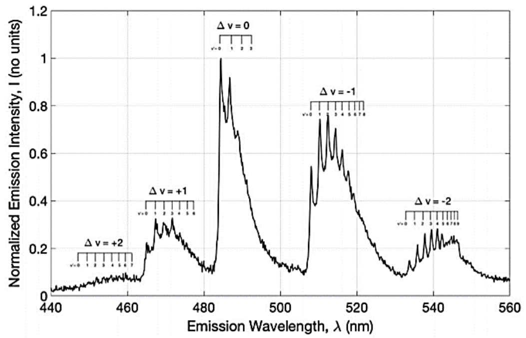
S.S. Harilal *et al., J. Anal. At. Spectrom.* **36**, 150 (2021).

Mission Relevance

- Aim to improve capabilities in the wide area environmental sampling of nuclear materials using optical detection techniques to
  - Detect undeclared nuclear fuel cycle activities
  - Monitor nuclear explosion events
- Plan to address this by developing optical signatures of Pu using Ce LPP surrogates
  - Ce is a common Pu surrogate due to similarities in electronic, thermodynamic, and chemical properties
- Requires an understanding of the
  - High-temperature gas-phase oxidation chemistry of Ce and Pu LPPs
  - Effect of plume hydrodynamics and mixing process with the surrounding environment on spectral signatures



S.S. Harilal *et al.*, *Anal. Chem.* **88**, (2016).

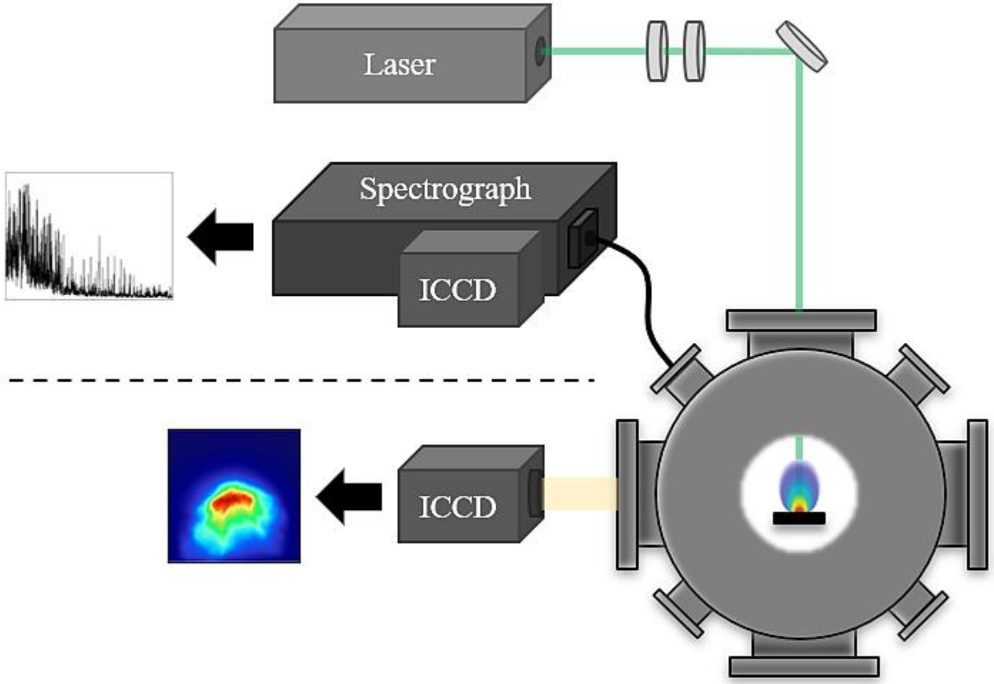


T.A. Van Woerkom *et al.*, *J. Opt. Soc. Am. B: Opt. Phys.* **35(10)**, 2018.

Technical Approach

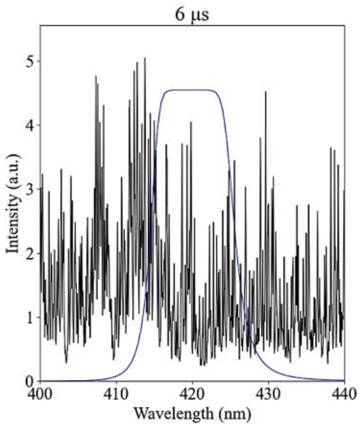
- Nanosecond laser ablation of cerium metal target
- Experiments:
  1) Laser-induced breakdown spectroscopy (LIBS)
  2) Time-resolved fast-gated imaging of total plasma emission
- Various ambient gases at 760 Torr

Experimental Setup

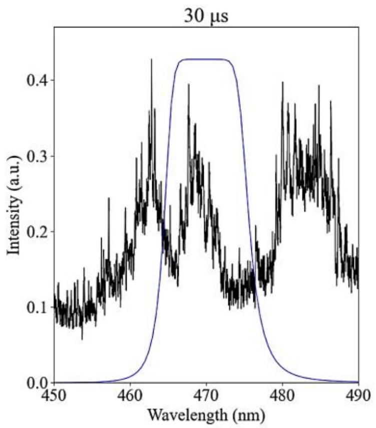


Filters used in the Imaging Experiments

Ce Emission
Bandpass Filter
$\lambda_0 = 420$ nm,
FWHM $= 10$ nm

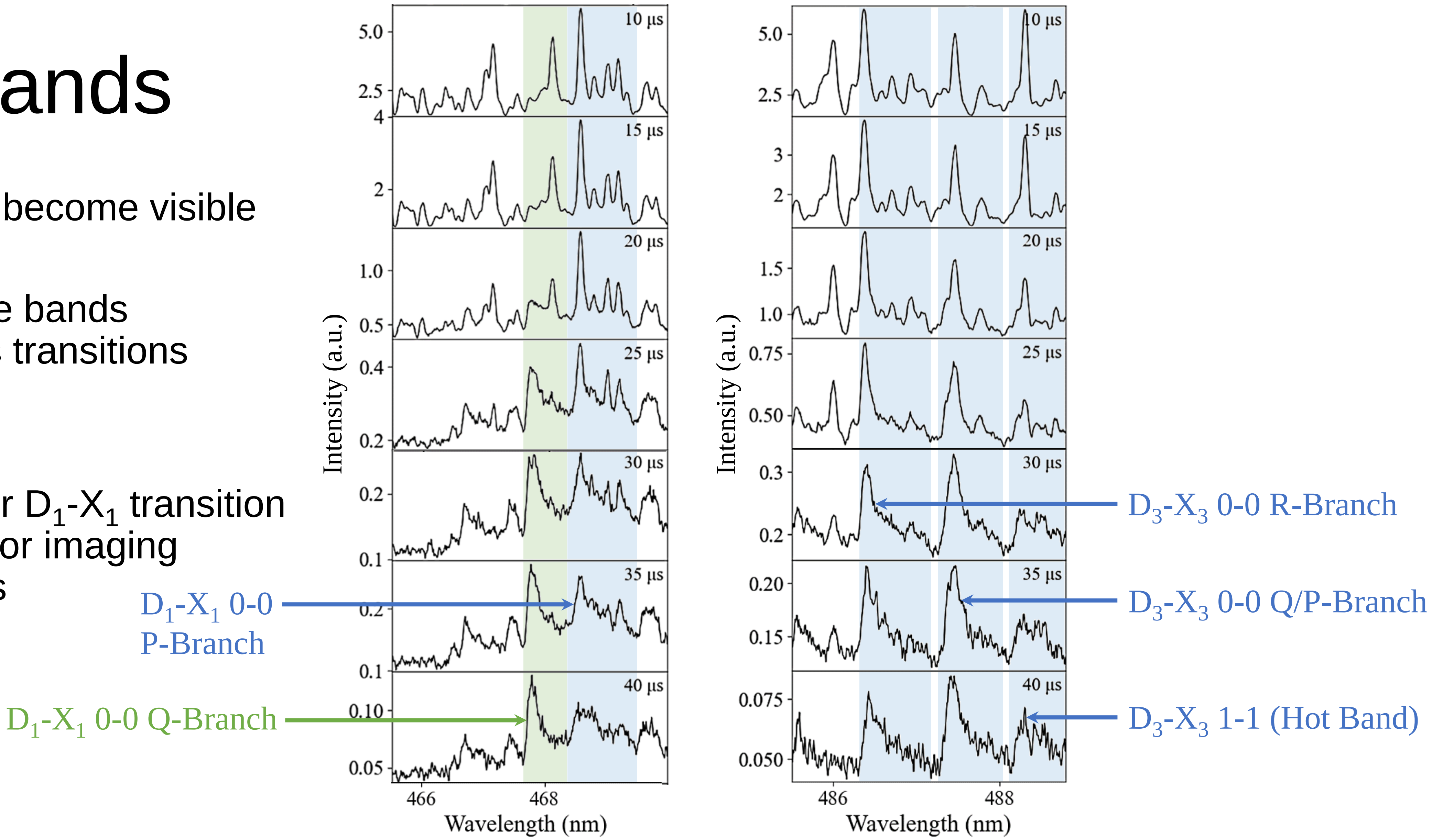


CeO Emission
Bandpass Filter
$\lambda_0 = 470$ nm,
FWHM $= 10$ nm

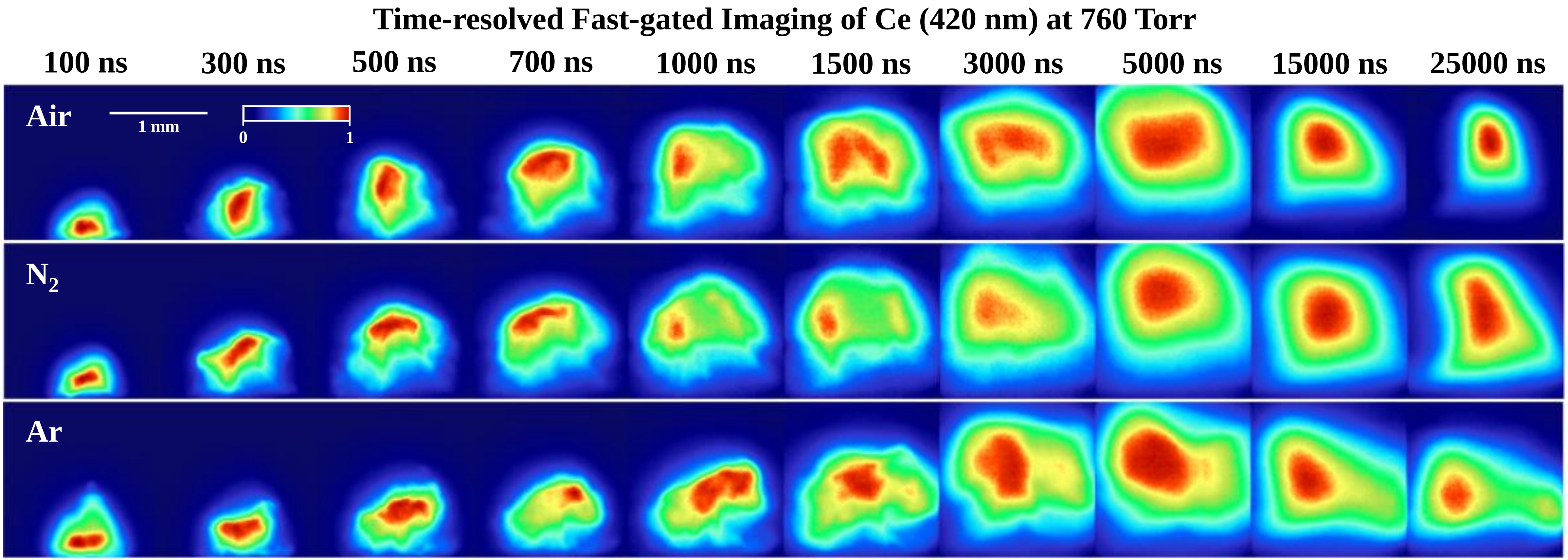


CeO Bands

- CeO bands become visible at ~20 µs
- Strong oxide bands identified as transitions
  - D_1-X_1
  - D_3-X_3
- Q-branch for D_1-X_1 transition is targeted for imaging experiments



Spatial Structure of Ce LPPs in Various Atmospheres



(1) Faster decay in total emission intensity in air compared to N_2 **(2)** Stronger emission in Argon due to higher gas density

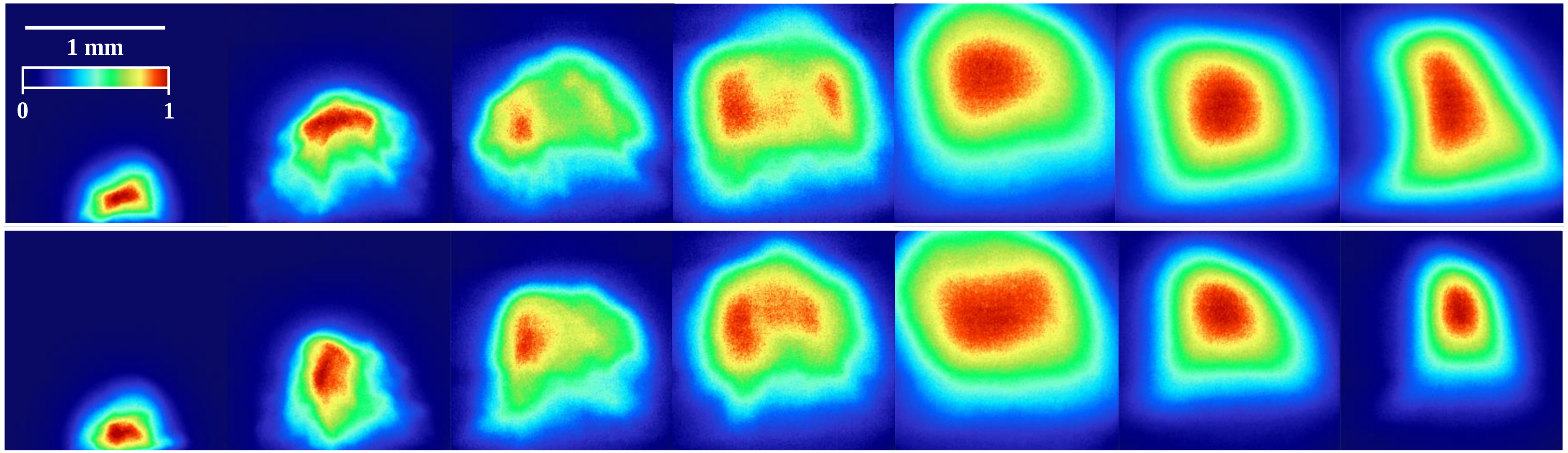
Effect of Oxidation on Total Plasma Emission

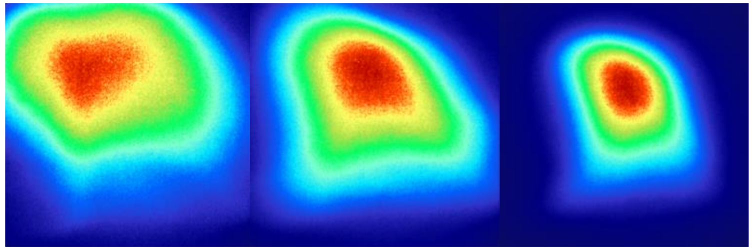
100 ns | 500 ns | 1000 ns | 2000 ns | 5000 ns | 15000 ns | 25000 ns

Atomic Ce Transitions in N_2

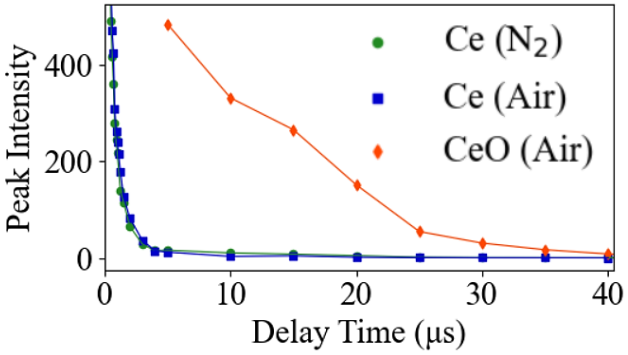
Atomic Ce Transitions in Air

Molecular CeO Transitions in Air





Includes atomic Ce emission due to large bandwidth of the bandpass filter



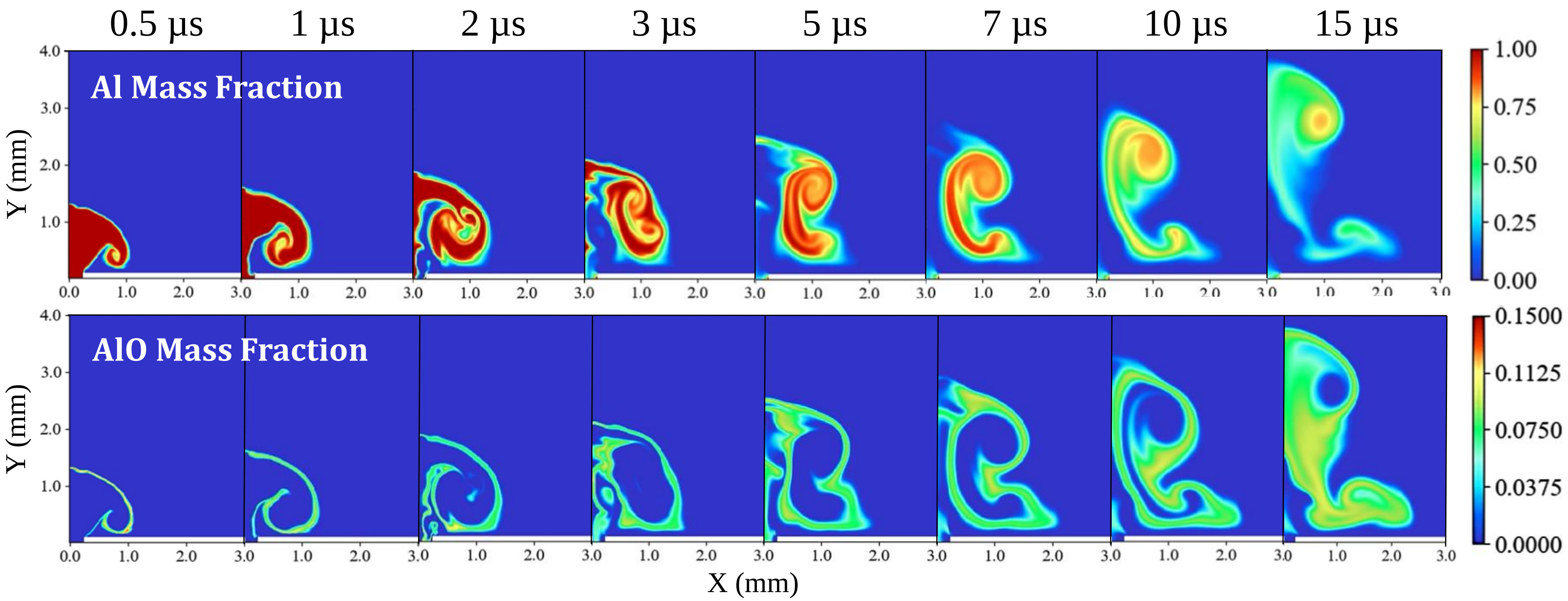
CeO emission is suppressed in the N_2 atmosphere

- Oxide layer on the cerium target plays minor role in observed combustion in the plasma plume
- Cleaning shots adequately remove surface oxidation

Expected Impact

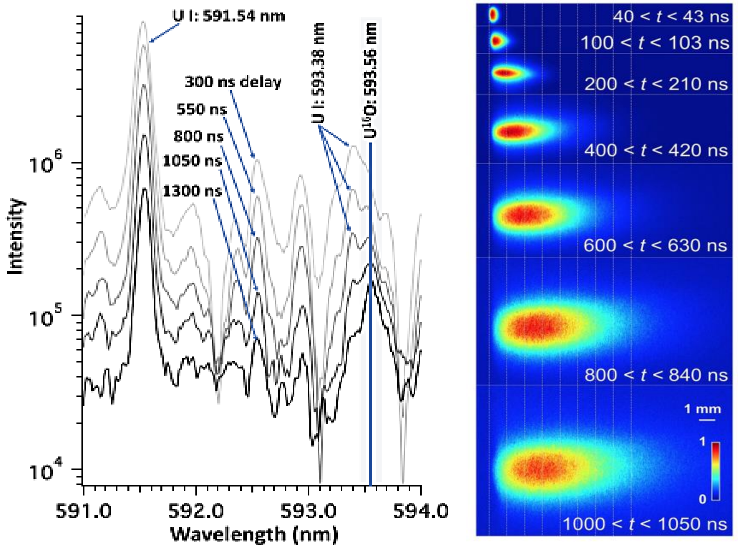
- Plan to elucidate plasma plume hydrodynamics and mixing process with the surrounding environment (e.g., air)
- Provide a more informed understanding of the high-temperature gas-phase oxidation chemistry of actinide LPPs

Multiphysics Simulation of Aluminum Fireball Combustion Chemistry in Air at 1 STP



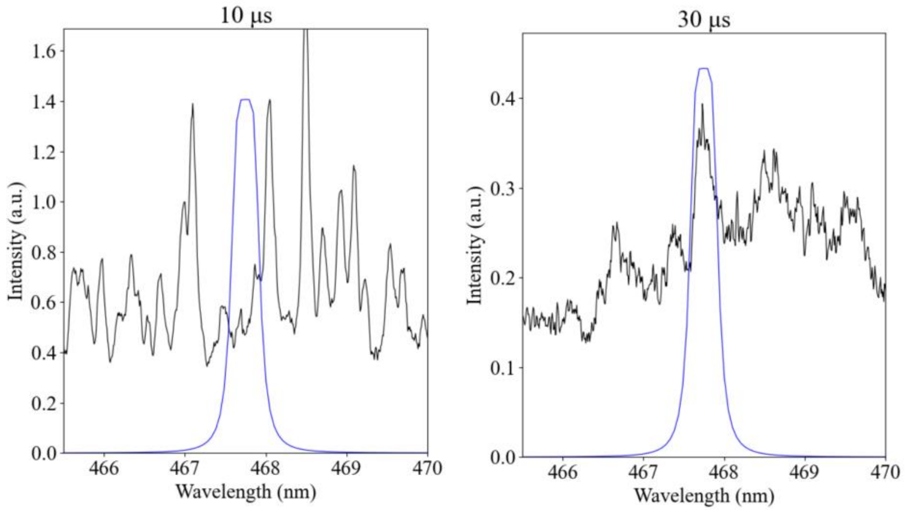
Next Steps

- Elucidate the effect of bulk pressure on spectroscopic Ce LPP signatures
- Elucidate the effect of oxygen concentration on the spatio-temporal evolution of Ce LPPs
  - Perform LIBS on Ce target in an inert atmosphere for various oxygen concentrations
  - Perform time-resolved fast-gated imaging of Ce LPP for same various conditions
    - Use custom bandpass filter with a narrower FWHM to mitigate collection of atomic emission concurrently with molecular emission



(left) D.G. Weisz *et al.*, *Appl. Phys. Lett.* **111**, 034101 (2017). (right) M. Burger *et al.*, *Phys. Plasmas* **26**, 093103 (2019).

Ce LPPs



Bandpass Filter
$\lambda_0 = 467.75$ nm,
FWHM $= 0.3$ nm

MTV Impact

- Internships:
  - Previous internships: NNL, DoD
  - Upcoming internship with DoD
- DoD SMART graduate fellowship
- Upcoming conferences:
  - MARC XII (April 3-8)
  - DTRA IIRM-URA Annual Technical Review (Estimated July; virtual)
  - INMM 63rd Annual Meeting (July 24-28; virtual)
  - SciX 2022 Conference (October 2-7)
- Research collaborations: **SRNL**, AFTAC, UM

Conclusion

- This work presents progress towards developing Pu optical signatures using Ce LPP surrogates
- CeO bands were identified to investigate the oxidation chemistry of Ce LPPs
- Time-resolved fast-gated imaging was performed to study the spatial structure and expansion dynamics of Ce LPPs
- Supports the NNSA mission by developing optical detection techniques to enable the standoff, in-field detection of actinides

Optical Science and Nonproliferation Group University of Florida

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