INL/MIS-23-75382-Revision-0

Xenon Detection Using Double-Pulse Laser-Induced Breakdown Spectroscopy

November 2023

Changing the World's Energy Future

Londrea Garrett, Milos Burger, Igor Jovanovic, Yunu Lee , Hyung-bin Kim, Sungyeol Choi , Piyush Sabharwall

Idaho National Laboratory

INL is a U.S. Department of Energy National Laboratory operated by Battelle Energy Alliance, LLC

INL/MIS-23-75382-Revision-0

Xenon Detection Using Double-Pulse Laser-Induced Breakdown Spectroscopy

Londrea Garrett, Milos Burger, Igor Jovanovic, Yunu Lee , Hyung-bin Kim, Sungyeol Choi , Piyush Sabharwall

November 2023

Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

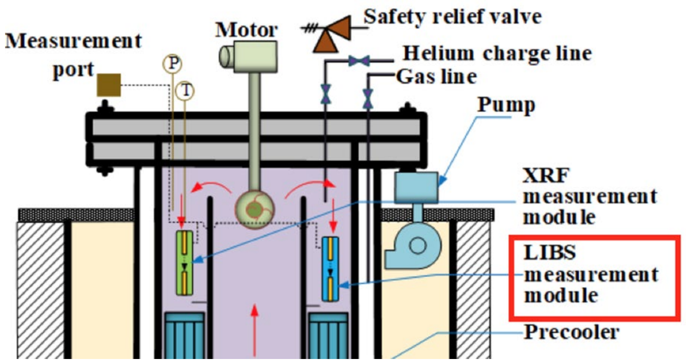
Xenon Detection Using Double-Pulse Laser-Induced Breakdown Spectroscopy

Londrea Garrett, Miloš Burger, Igor Jovanovic – *University of Michigan*
Yunu Lee – *Korea Advanced Institute of Science and Technology*
Hyung-bin Kim, Sungyeol Choi – *Seoul National University*
Piyush Sabharwall – *Idaho National Laboratory*

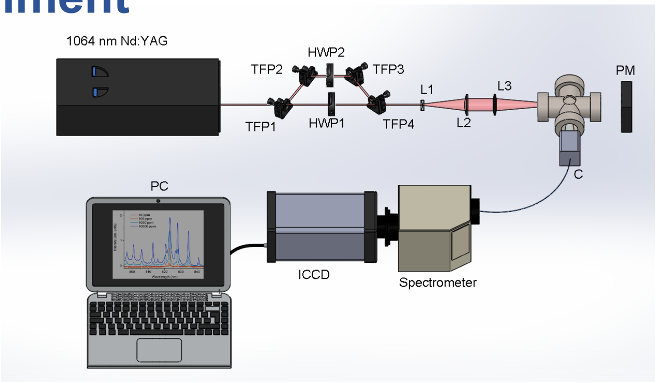
UNIVERSITY OF MICHIGAN

Overview

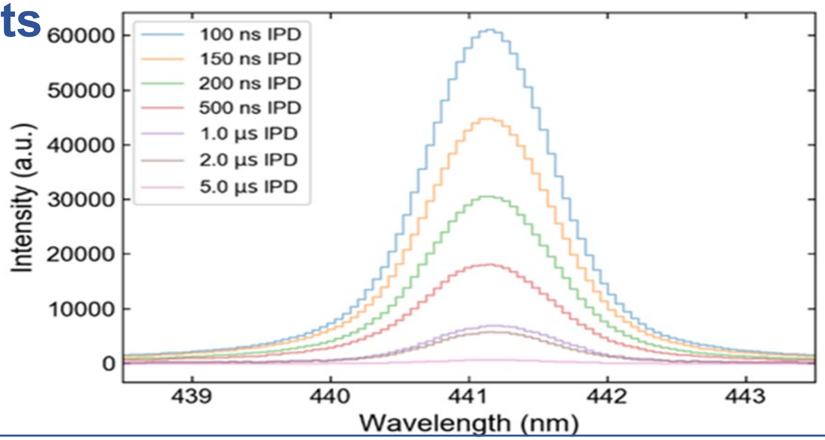
Background



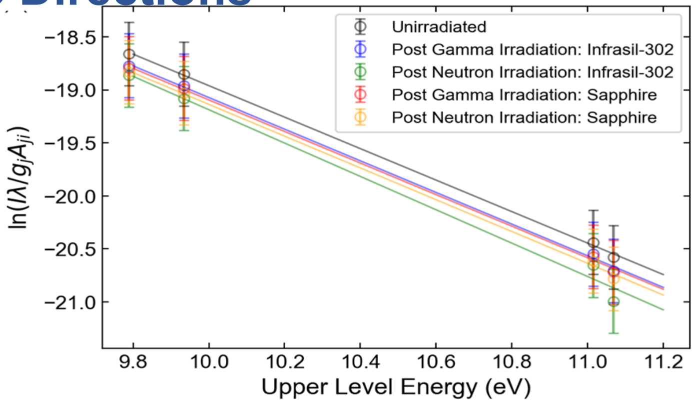
Experiment



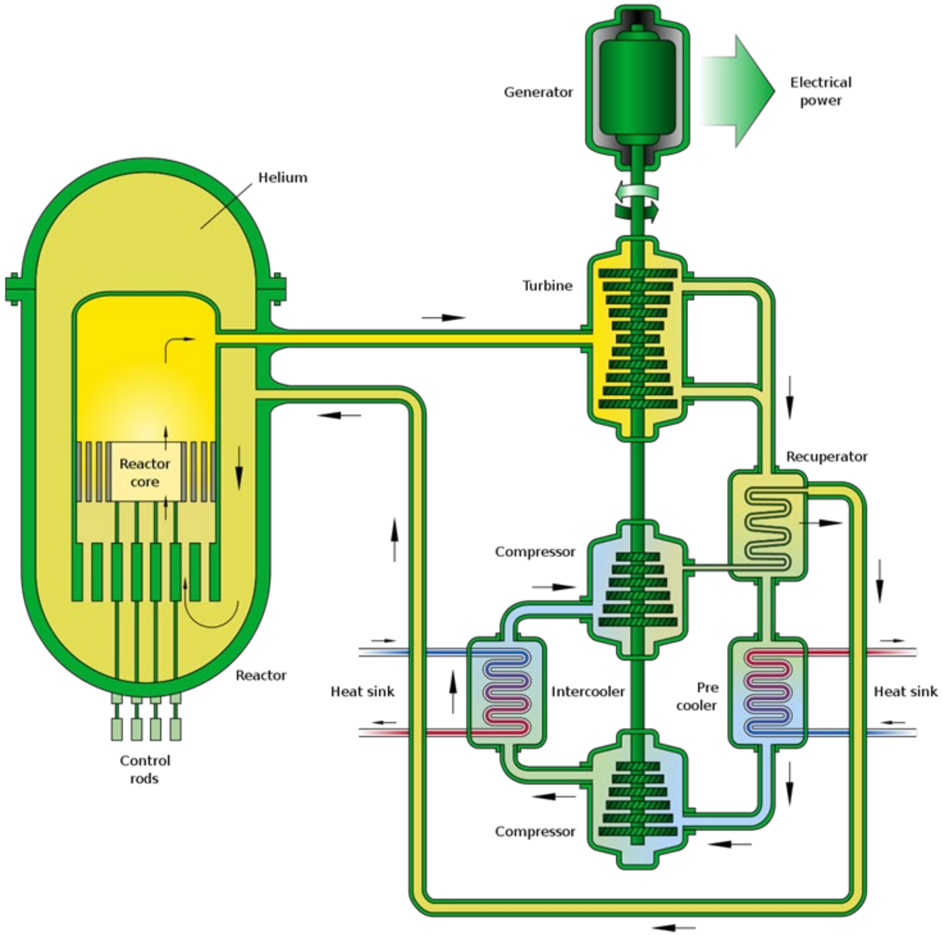
Results



Future Directions



Generation IV helium-cooled fast reactors will benefit from *in-situ* diagnostics

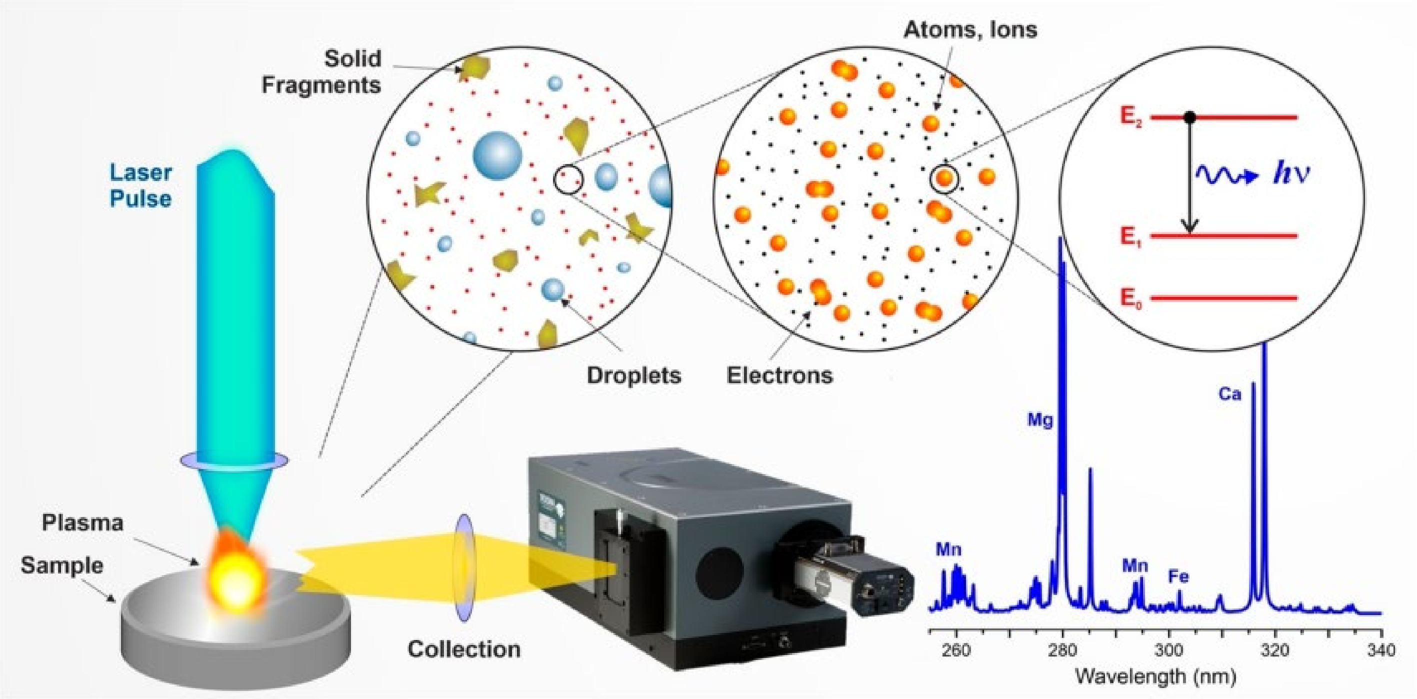


gen-4.org (2022)

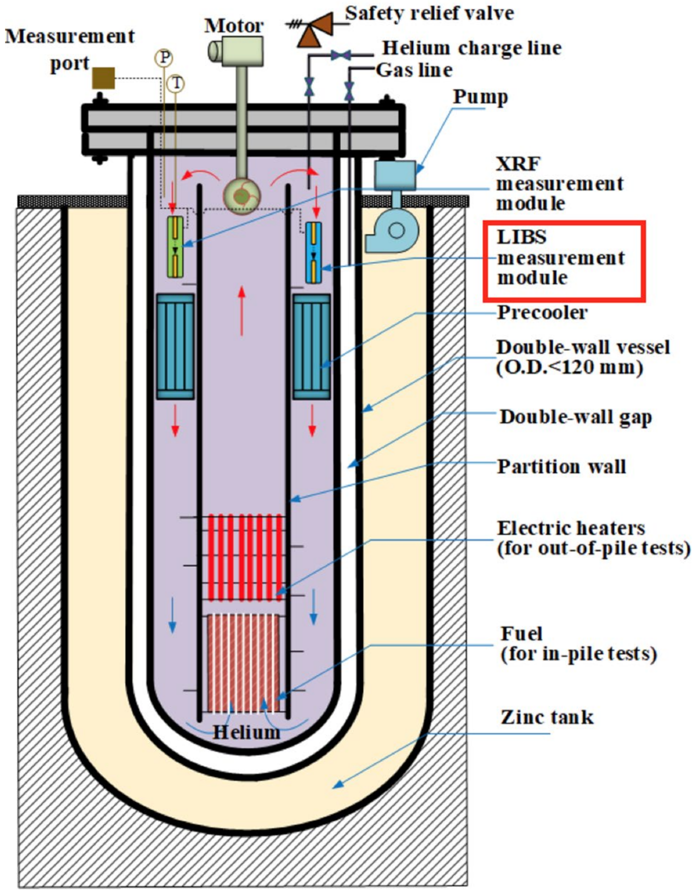
Challenges facing instrumentation include:

- High temperature
- High pressure
- High ionizing radiation flux
- Limited understanding of new fuel and construction materials

Laser-induced breakdown spectroscopy (LIBS) has been proposed for reactor fuel-failure monitoring



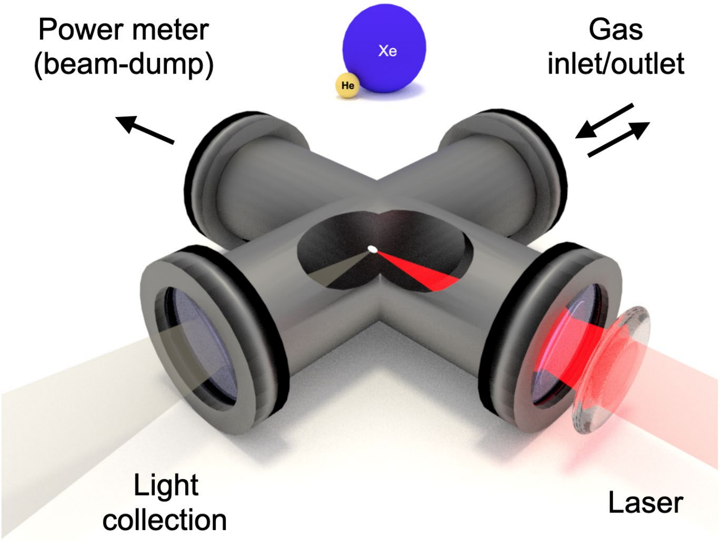
Gardette et al. *Anal. Chem*, **95** (2023). pp 46-69

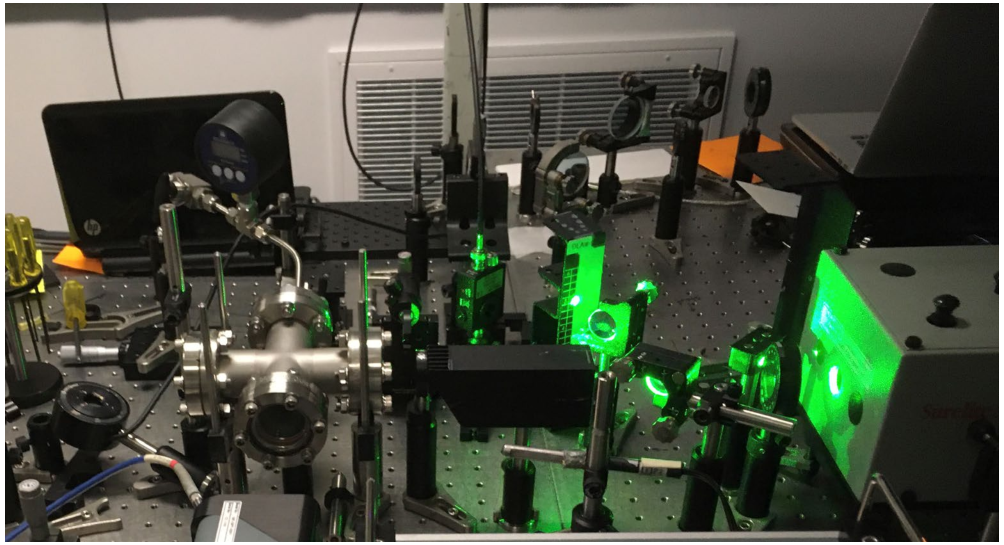


Sabharwall et al. Idaho National Laboratory (2020)

We developed a room-temperature testbed operated at atmospheric pressure

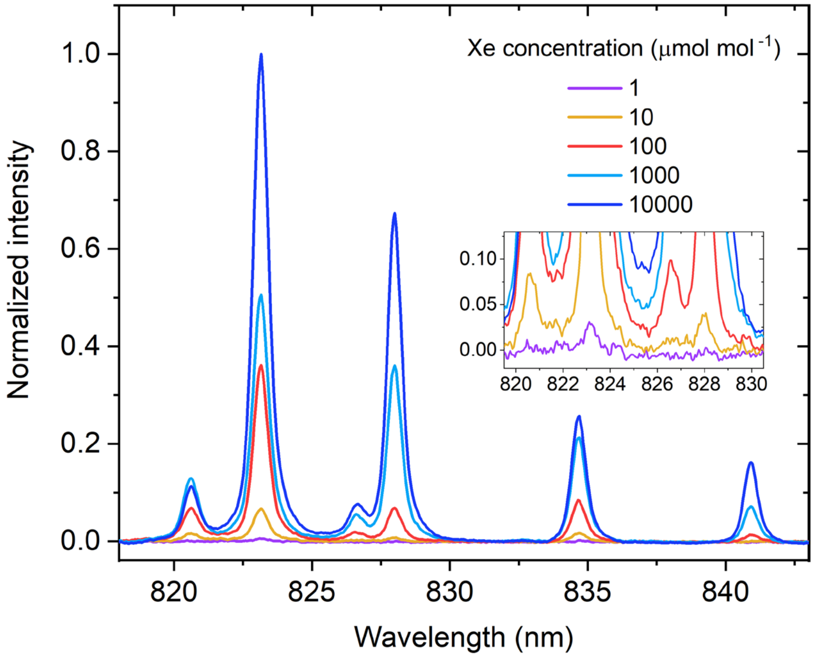
- Experimental goal: determine LIBS LOD for Xe in ambient He to represent a gas-cooled reactor analyte
- Measurements taken using a 1064 nm Nd:YAG laser with a pulse energy of 250 mJ
- Analysis based on the 823.16 nm Xe I spectral line



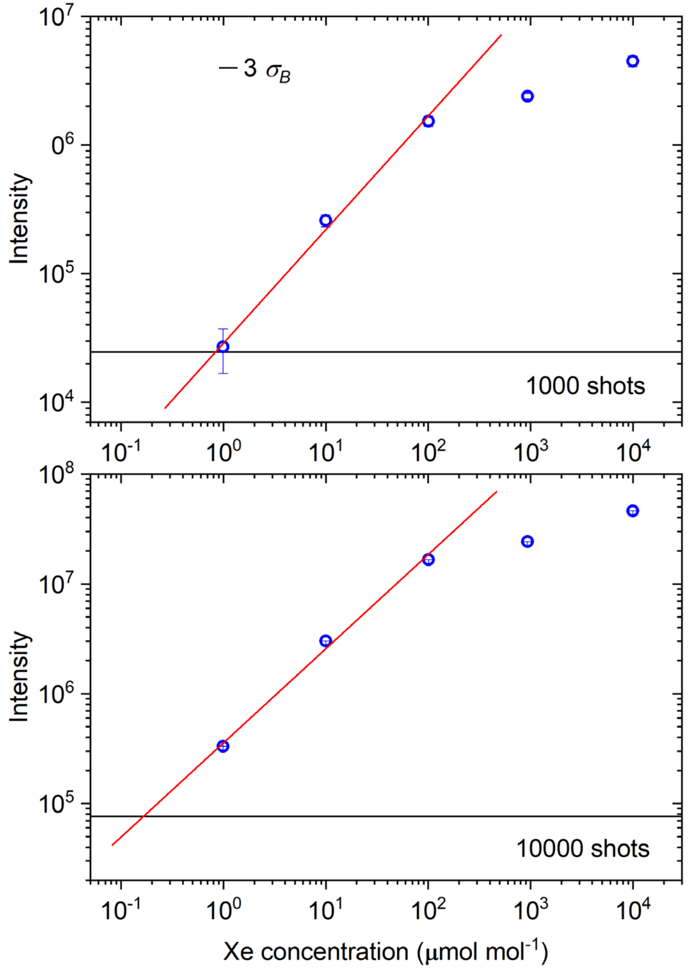


Previous work demonstrated 1-ppm sensitivity for Xe in He based on 10^4 laser shots

- Appropriate for detection of accident conditions
- Standard Xe concentrations expected to be between 9 to 30 ppb during reactor operation
- Gas chromatography and ICP-MS demonstrate limits of detection on the order of tens of ppb

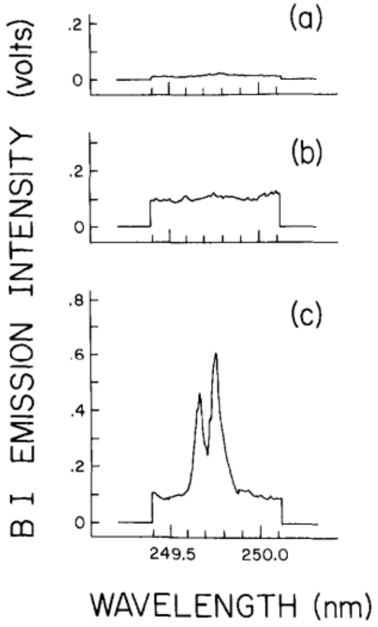


Burger et al., *J. Anal. At. Spectrom.* **36**, 824-828 (2021)

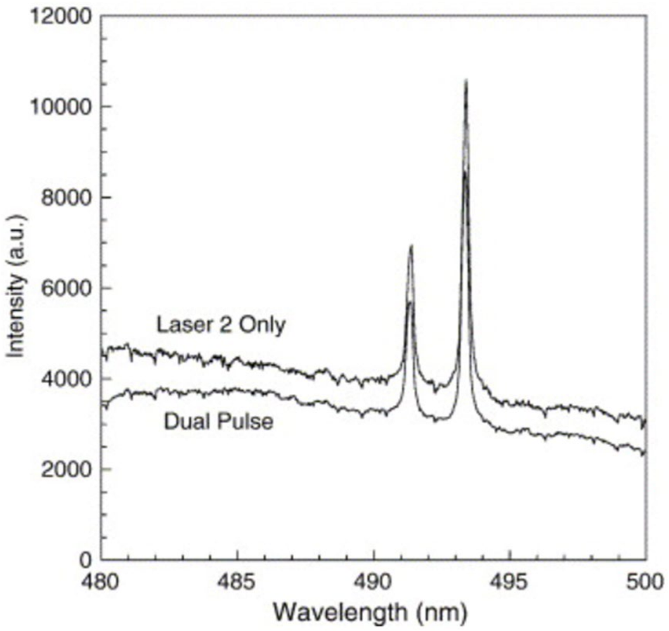


We demonstrate that double-pulse (DP) LIBS improves the sensitivity for Xe detection

- DP-LIBS has demonstrated a signal enhancement of 1–2 order of magnitude for solid analytes
- Previous studies note little improvement for gaseous samples
- We base our analysis on the 441.84 nm Xe I line to avoid interference from nitrogen spectral lines

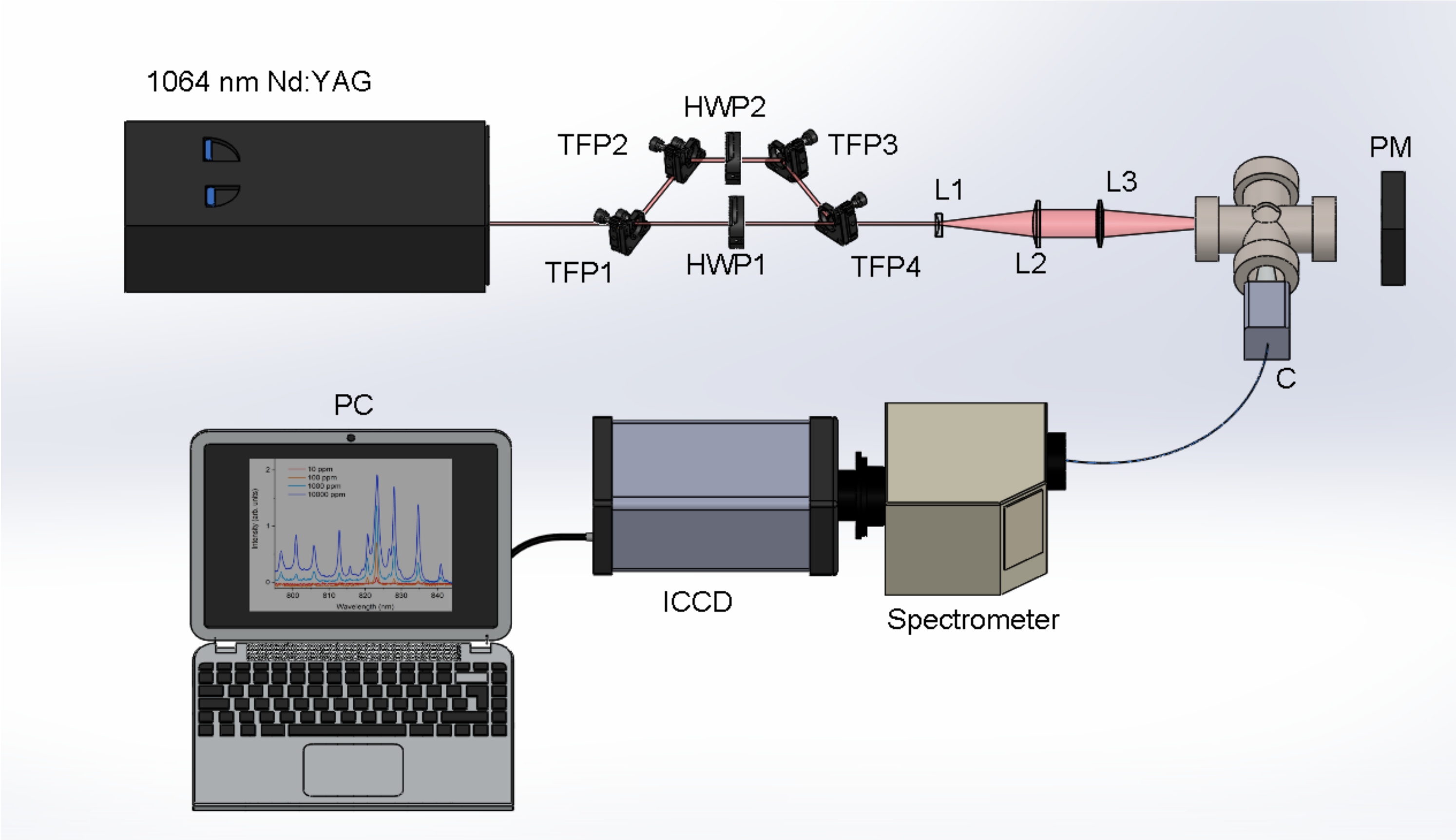


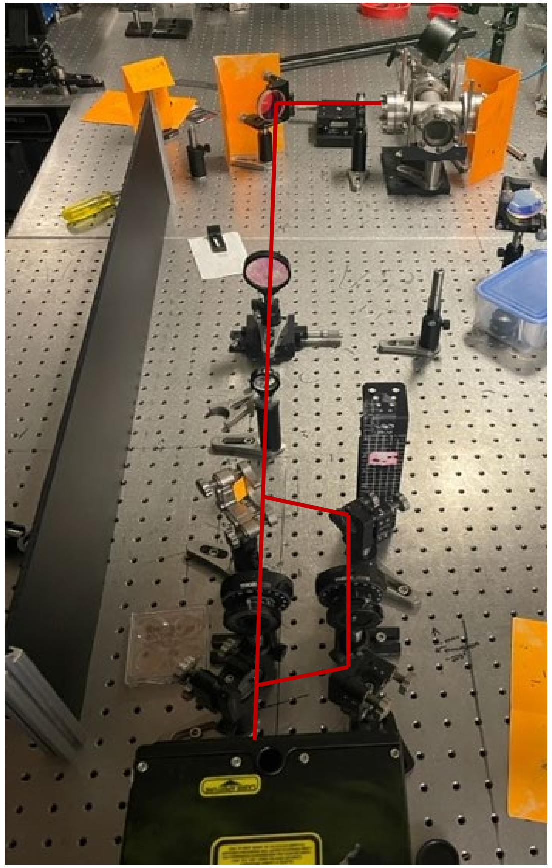
Cremers et al. *Appl. Spectrosc.*, **38**, 721-729 (1984).



Windom et al. *Spectrochim. Acta Part B At. Spectrosc*, **61**, 788-796 (2006).

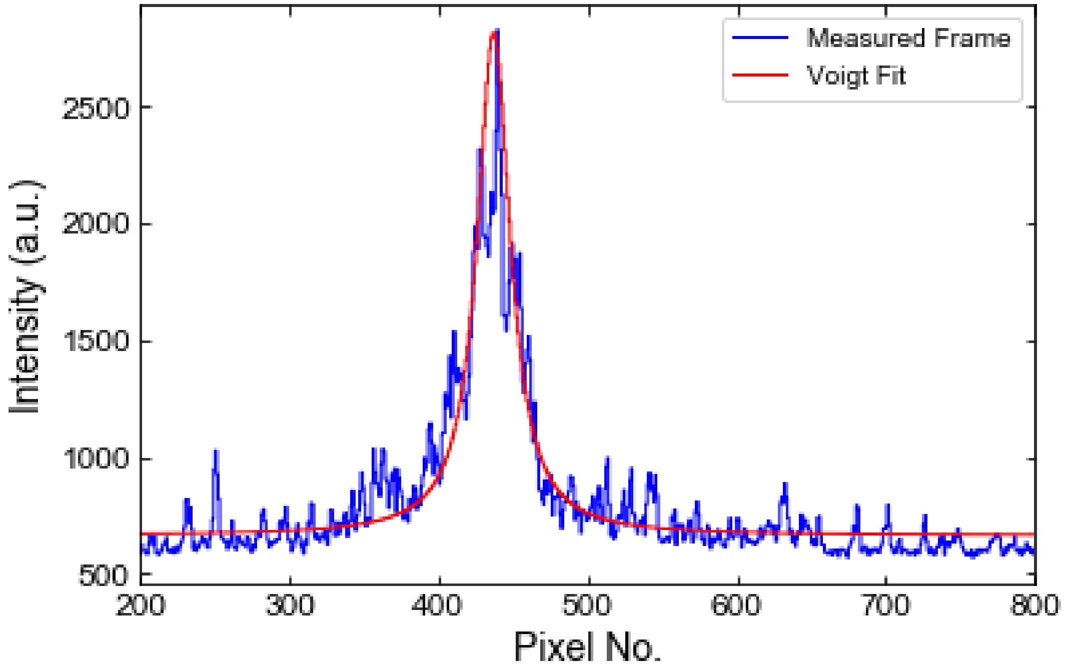
A laboratory setup was established to study double-pulse LIBS sensitivity for Xe detection





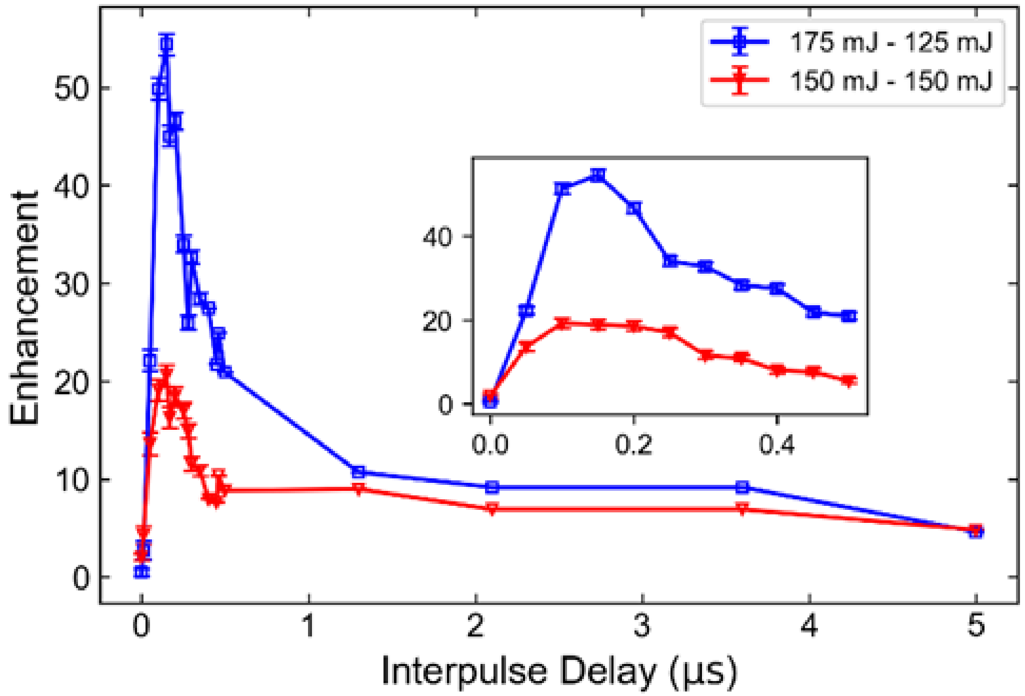
An analytical framework for spectral analysis is used to quantify the enhancement

- Enhancement: $$E = \frac{I_{DP}}{I_{SP}}$$
- Voigt Fit: $$V(\lambda;\sigma,\gamma) = I_0 \int_{-\infty}^{\infty} G(\lambda;\sigma) L(\lambda - \lambda';\gamma) d\lambda'$$
$$= \frac{I_0 Re\left[e^{z^2} erfc(-iz)\right]}{\sigma\sqrt{2\pi}}$$
$$z = \frac{\lambda + i\gamma}{\sigma\sqrt{2}}$$
- SNR: $$SNR = \frac{I_0}{\sqrt{\frac{\Sigma(X_i - <X>)^2}{N}}} \approx aN^b$$

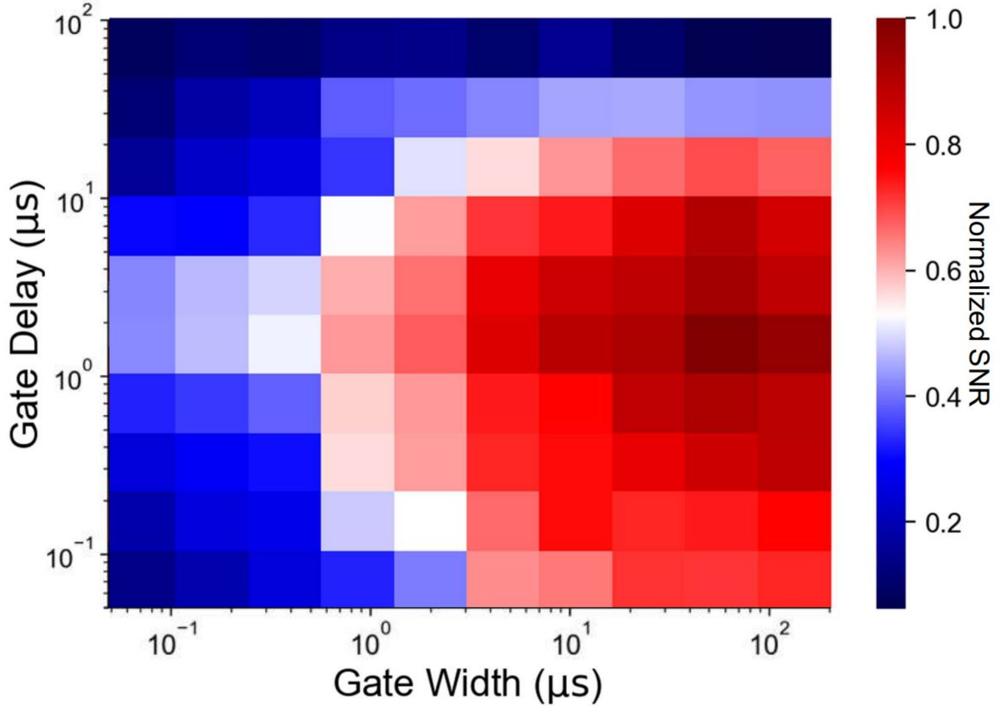


The large parameter space requires several optimizations

Enhancement vs. Interpulse Delay

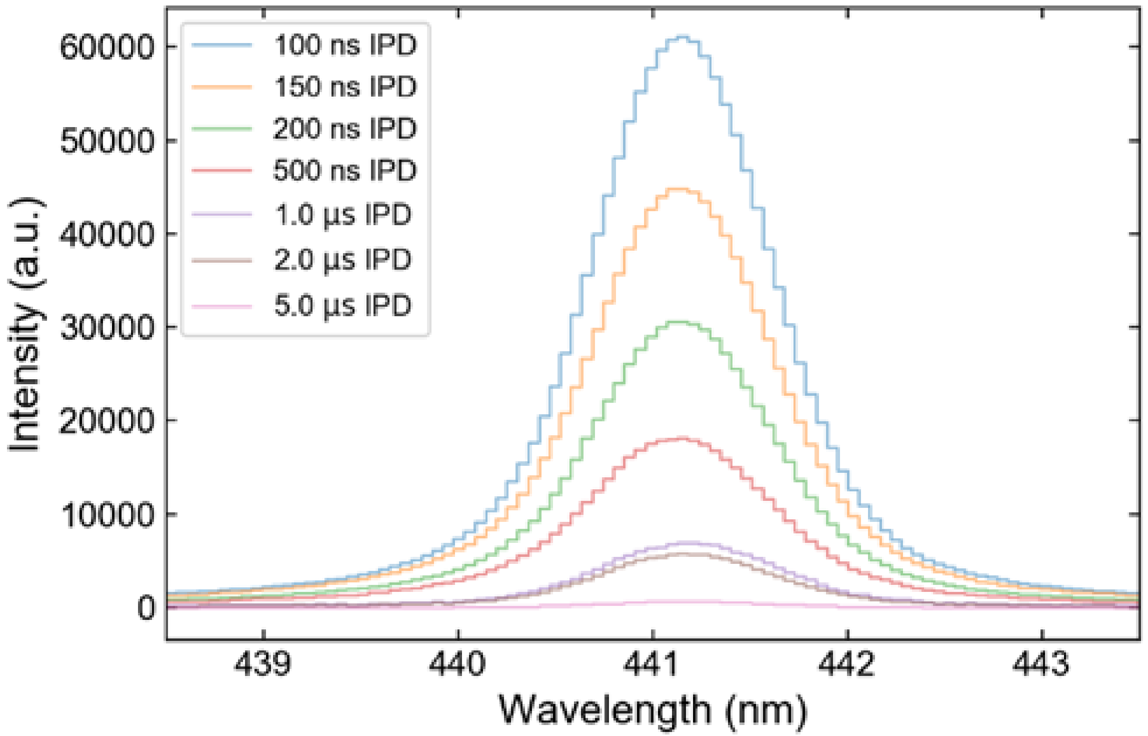


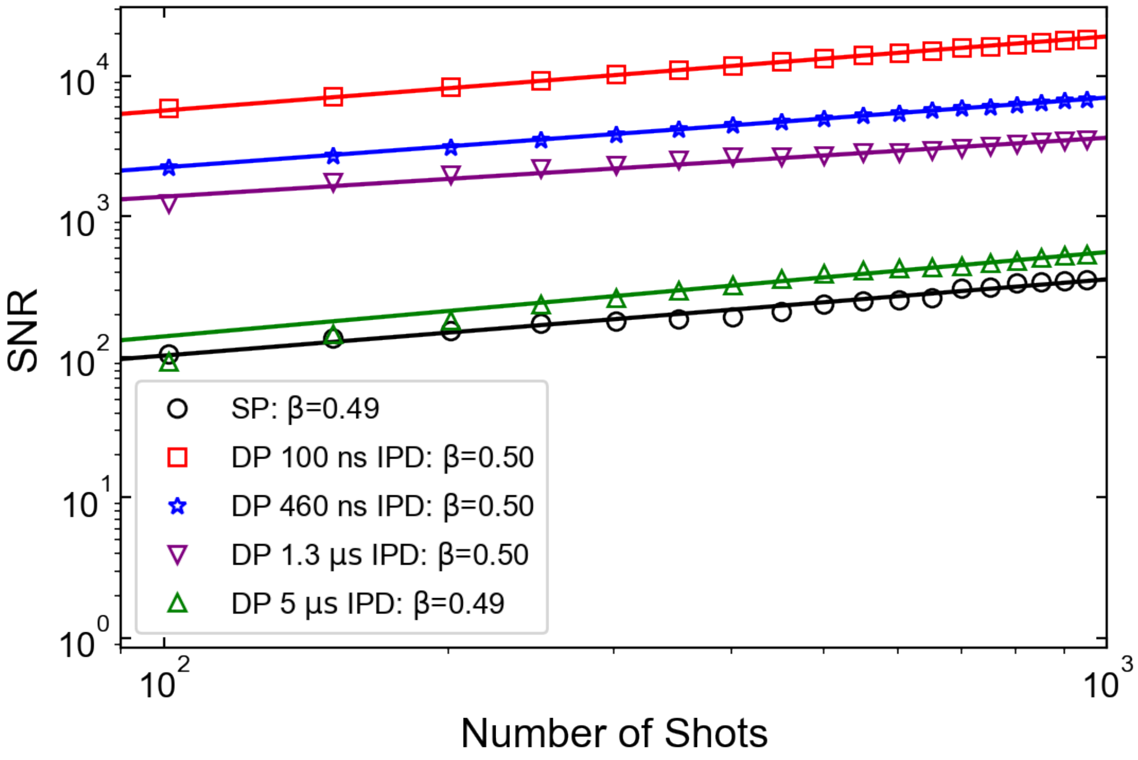
SNR vs. ICCD Recording Parameter



A 175 mJ – 125 mJ pulse energy combination, 150 ns interpulse delay, 1 μs gate delay, and 100 μs gate width were selected

A ~50x enhancement is observed in the 1 ppm signal at optimal conditions



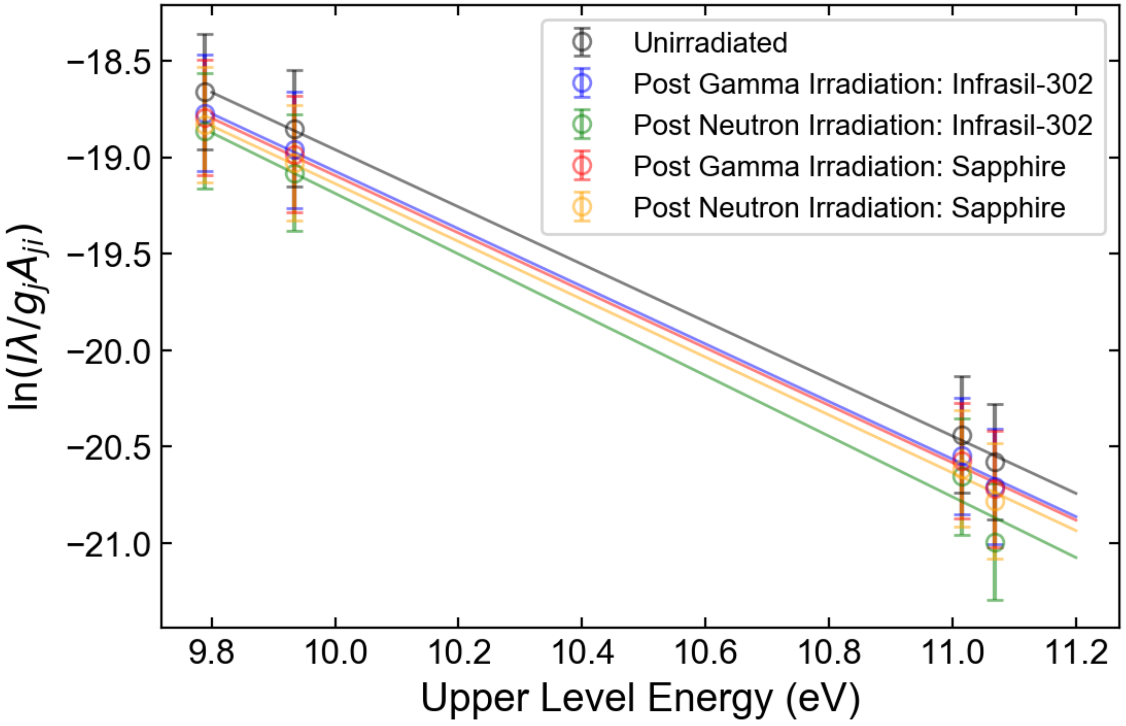


Preliminary DP-LIBS LOD Estimate: 4 ppb

Future work will aim to determine the optimal spectral region for measurement and the underlying physics of enhancement

- Using the 834.68 nm line will allow for comparison with nearby lines

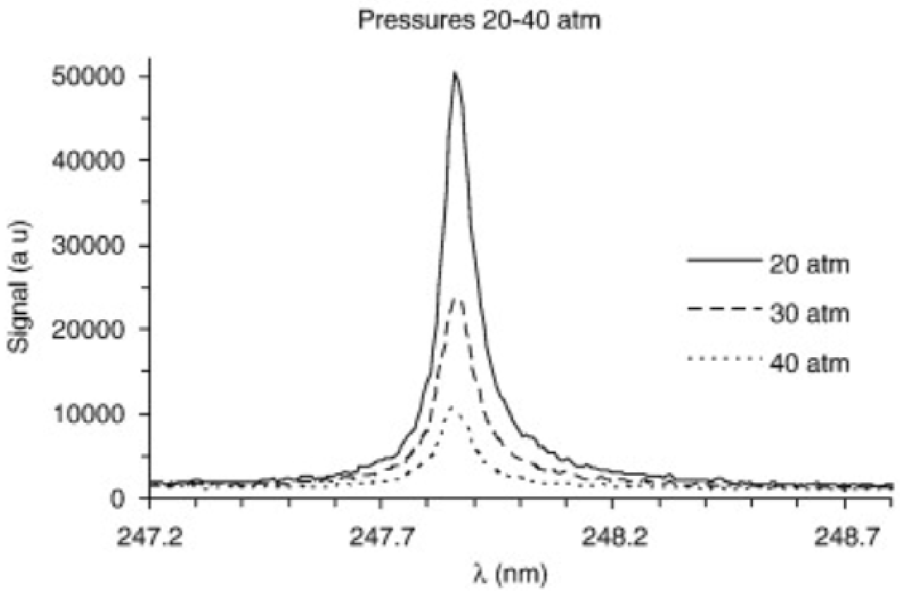
- Possible enhancement mechanisms:
  - Increased temperature and density
  - Longer emission lifetimes
  - Rarefication of the gaseous environment



Burger et al., *J. Anal. At. Spectrom.* **36**, 824-828 (2021).
Garrett et al., *Sensors,* **23 (2)**, (2023)

Studies will be needed to address the effects of environment temperature and pressure on spectral features

HTGR operational conditions are on the order of 850º C and 13 MPa

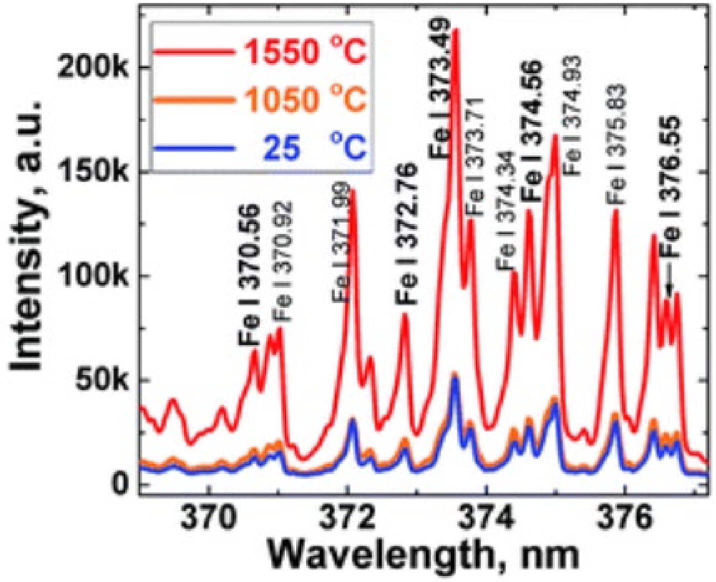


Increased Pressure

↓

Spectral Line Broadening

Vors et al., *Spectrochim. Acta Part B At. Spectrosc.* **63**, 1189-1204 (2008)



Increased Temperature

↓

Higher Signal Intensity

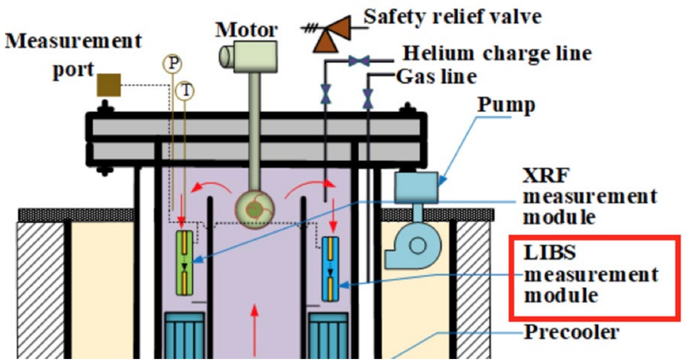
Lednev et al., *J. Anal. At. Spectrom,.* **34**, 607-615 (2019)

Acknowledgements

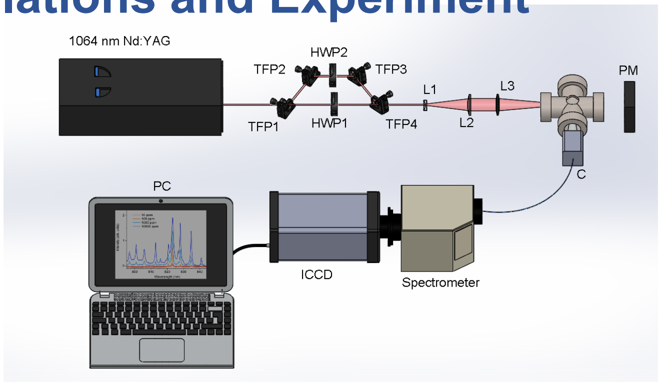
This research was performed under appointment to the Nuclear Nonproliferation International Safeguards Fellowship Program sponsored by the Department of Energy, National Nuclear Security Administration's Office of International Nuclear Safeguards (NA-241)

Summary

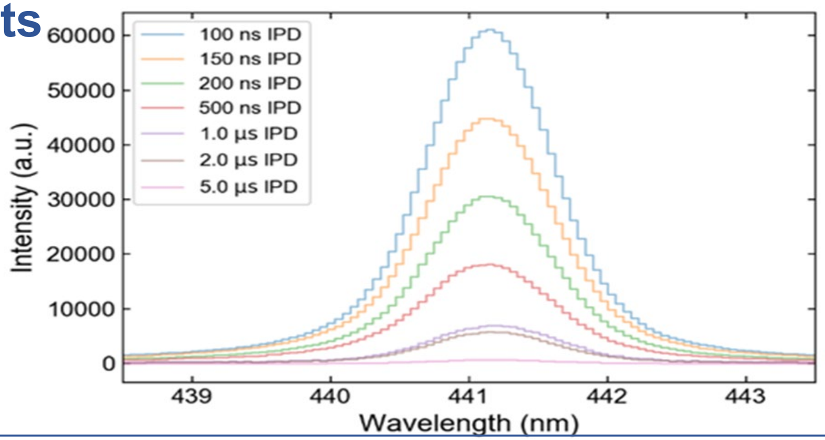
Background



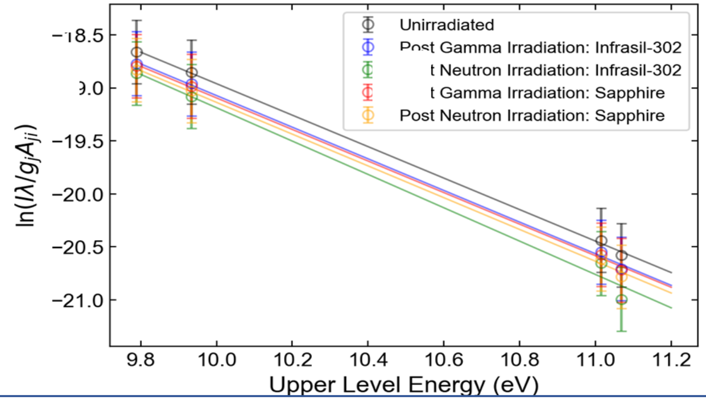
Calculations and Experiment



Results



Future Directions



Questions?