



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



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The logo for the ANS Winter 23 Conference and Expo. It features a blue circular icon with a white atomic symbol on the left. To its right, the text "ANS® Winter" is in blue, "Conference and Expo" is in black, and a large red "23" is on the far right. The background is a photograph of a modern, brightly lit hallway with a grid ceiling and people walking.

ANS® Winter 23 Conference and Expo

**MAINTAINING the
MOMENTUM**

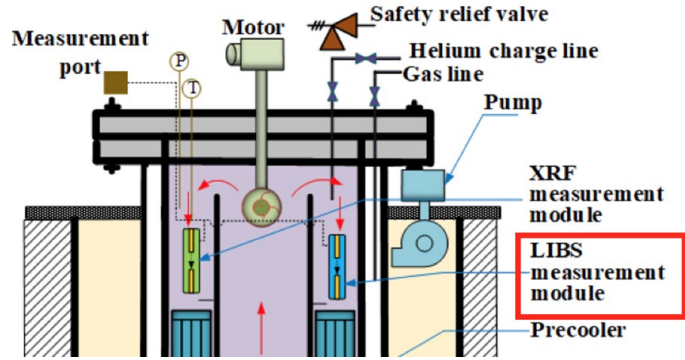
Xenon Detection Using Double-Pulse Laser-Induced Breakdown Spectroscopy

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*

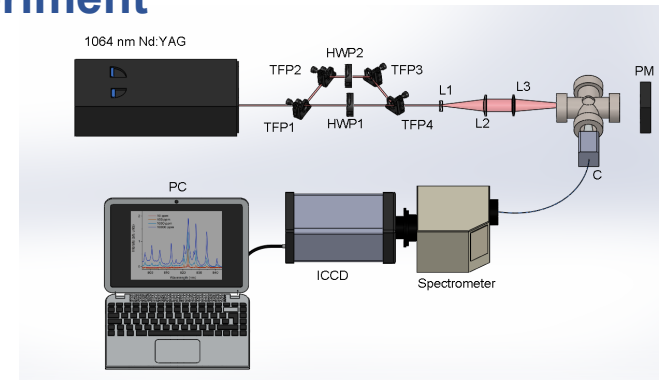


Overview

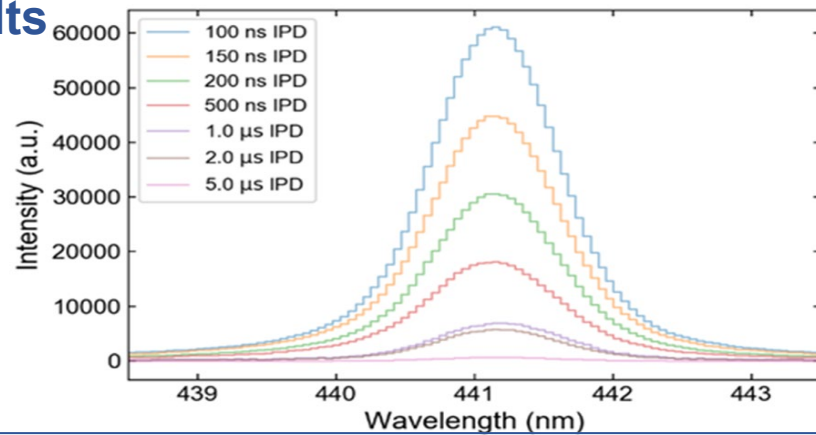
Background



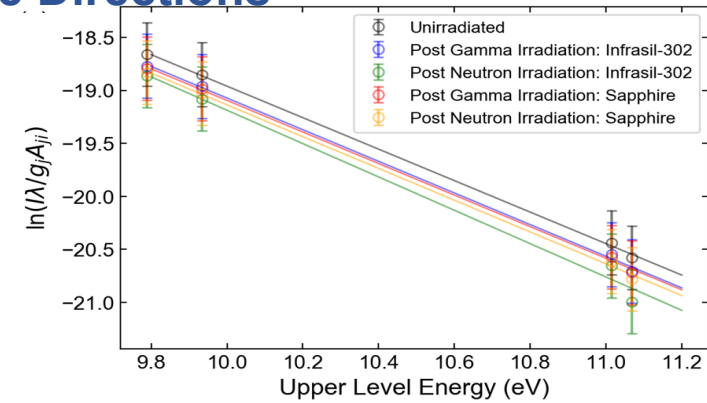
Experiment



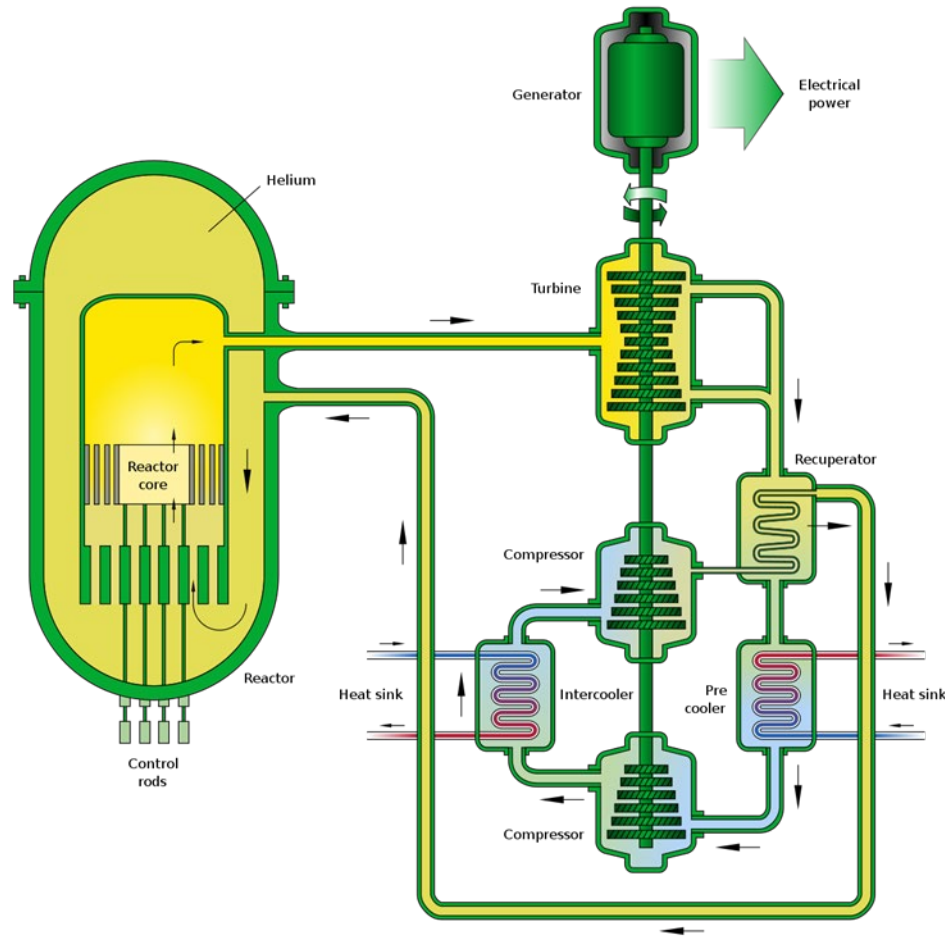
Results



Future Directions



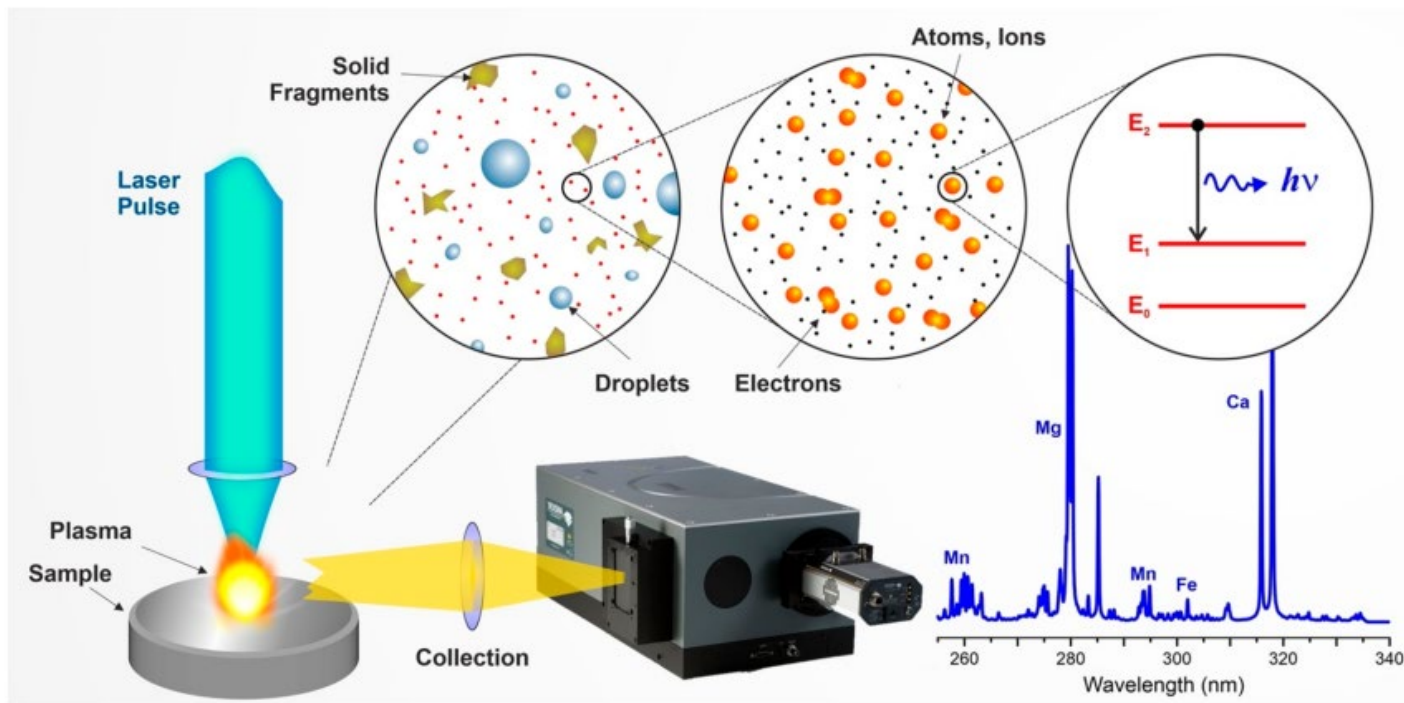
Generation IV helium-cooled fast reactors will benefit from *in-situ* diagnostics



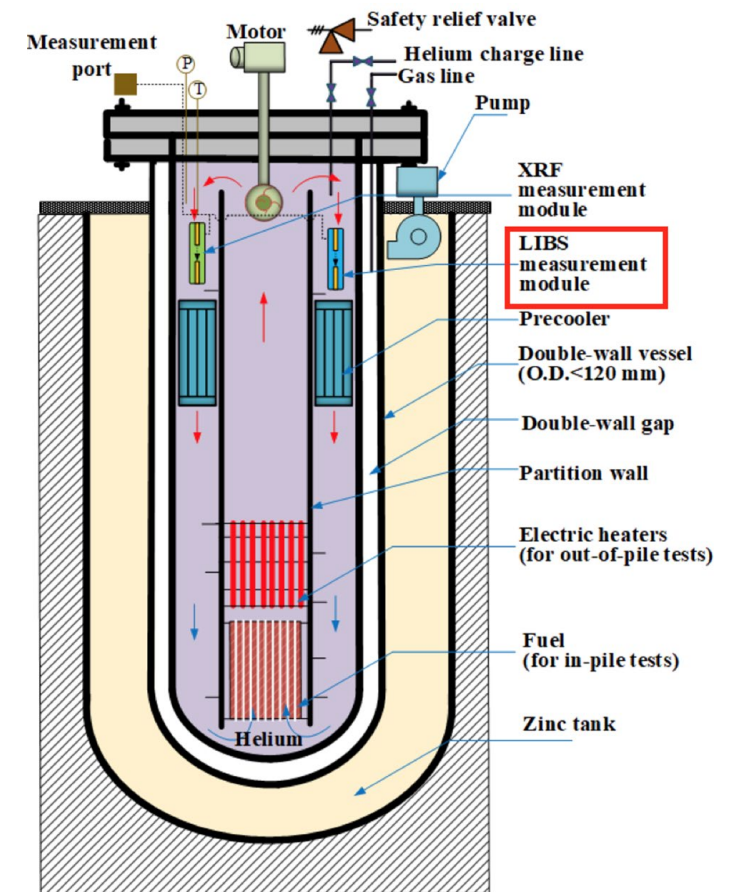
Challenges facing instrumentation include:

- 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



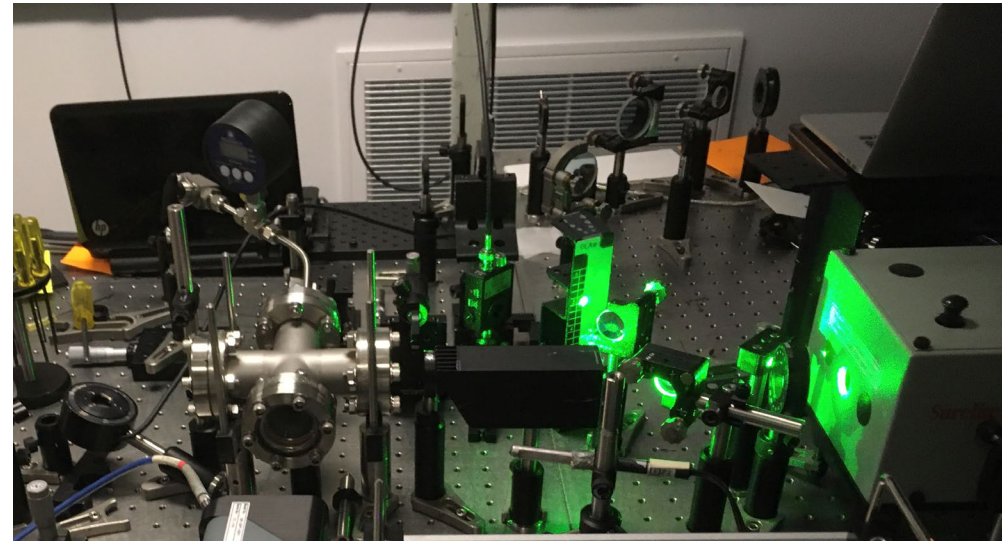
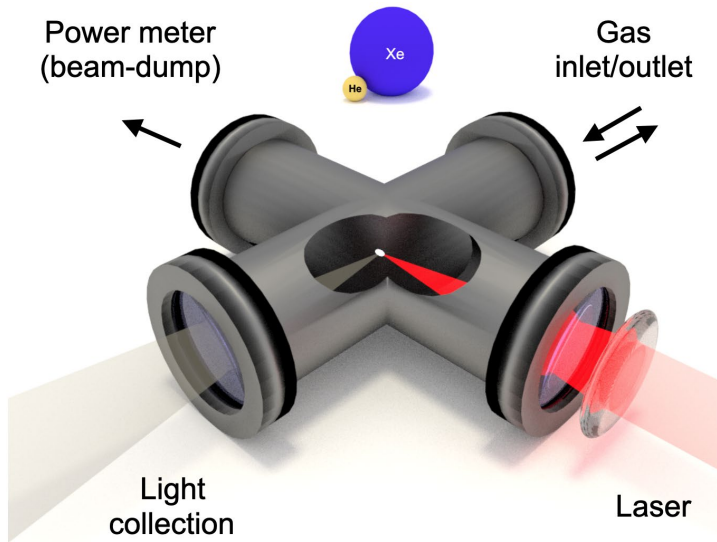
Gardette et al. *Anal. Chem*, **95** (2023). pp 46-69



Sabharwall et al. Idaho National Laboratory (2020)

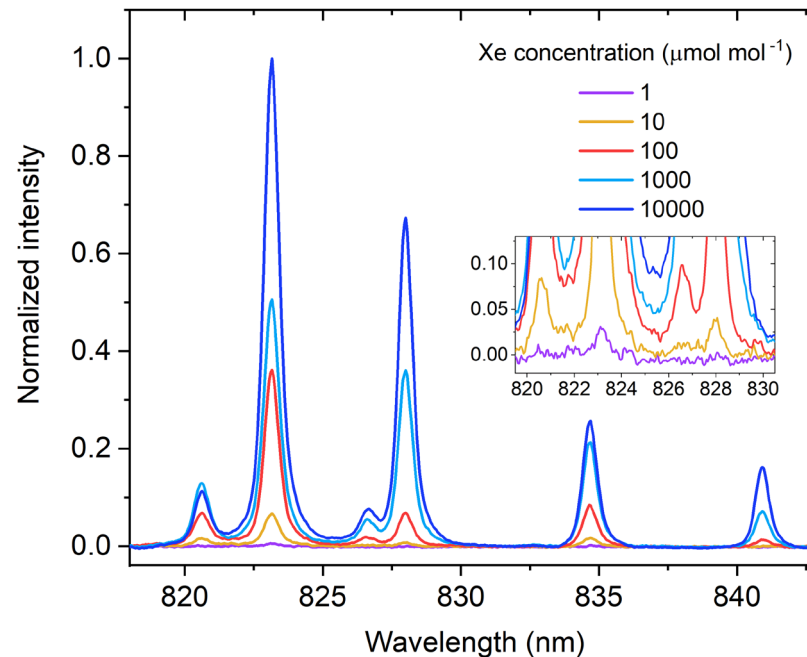
We developed a room-temperature testbed operated at atmospheric pressure

- 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

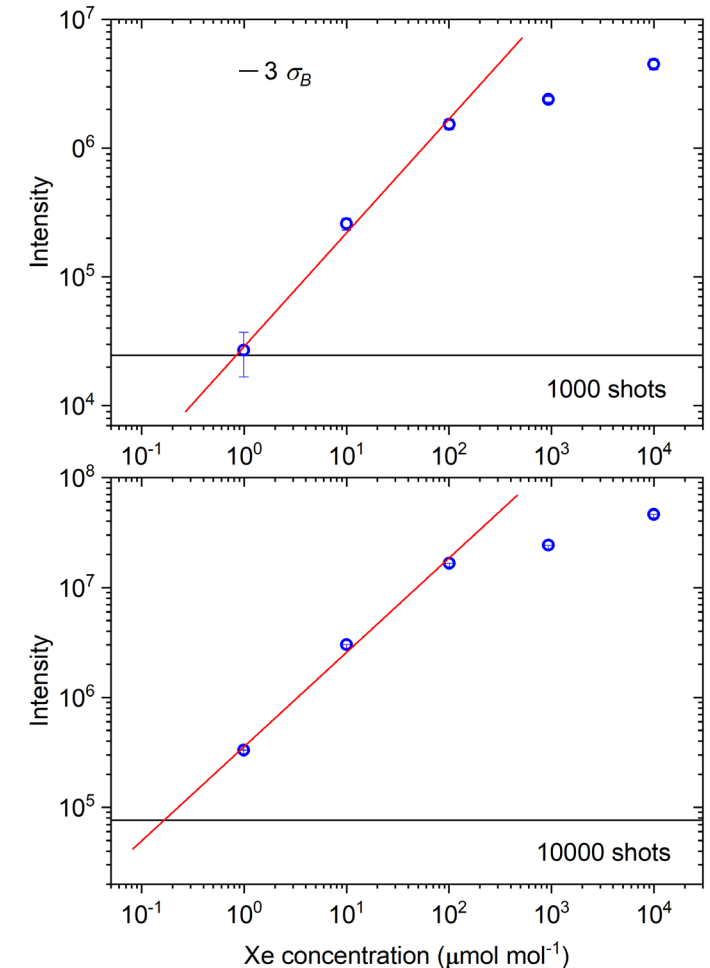


Previous work demonstrated 1-ppm sensitivity for Xe in He based on 10^4 laser shots

- 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

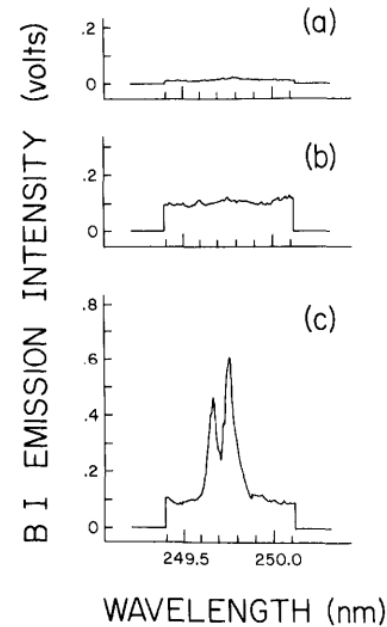


Burger et al., *J. Anal. At. Spectrom.* **36**, 824-828 (2021)

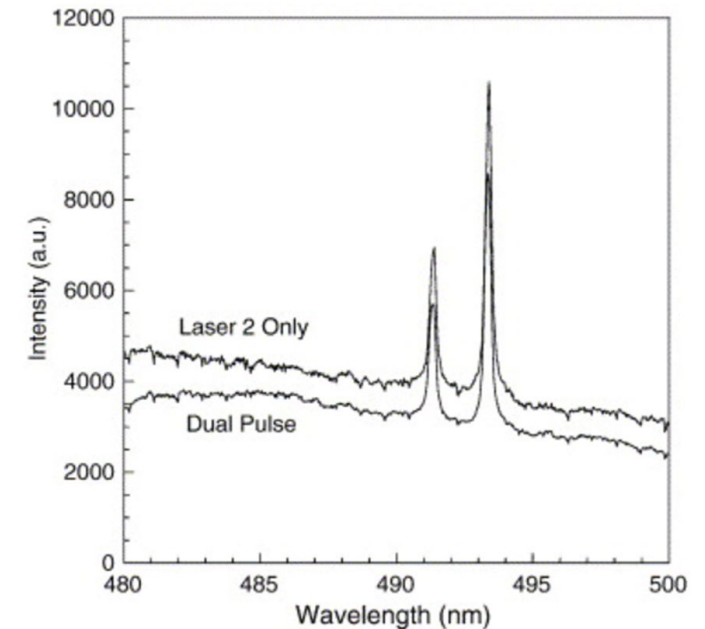


We demonstrate that double-pulse (DP) LIBS improves the sensitivity for Xe detection

- 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

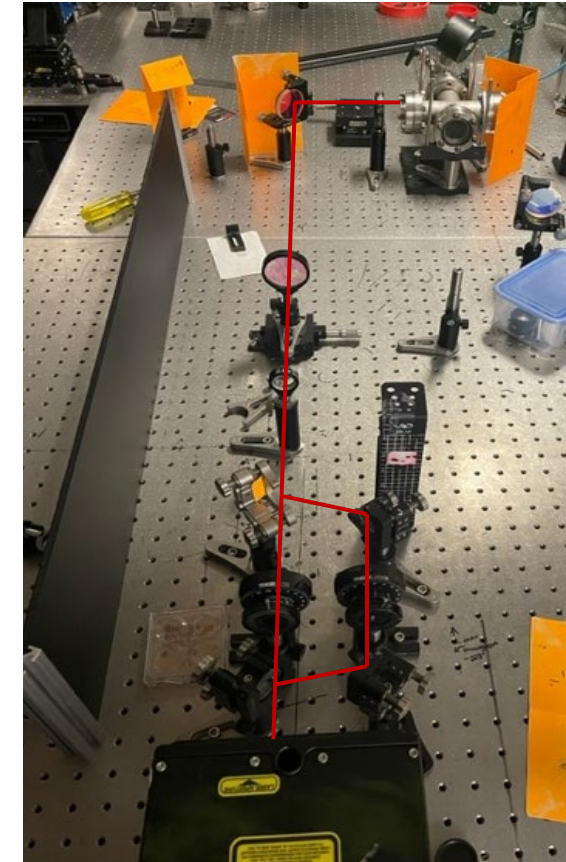
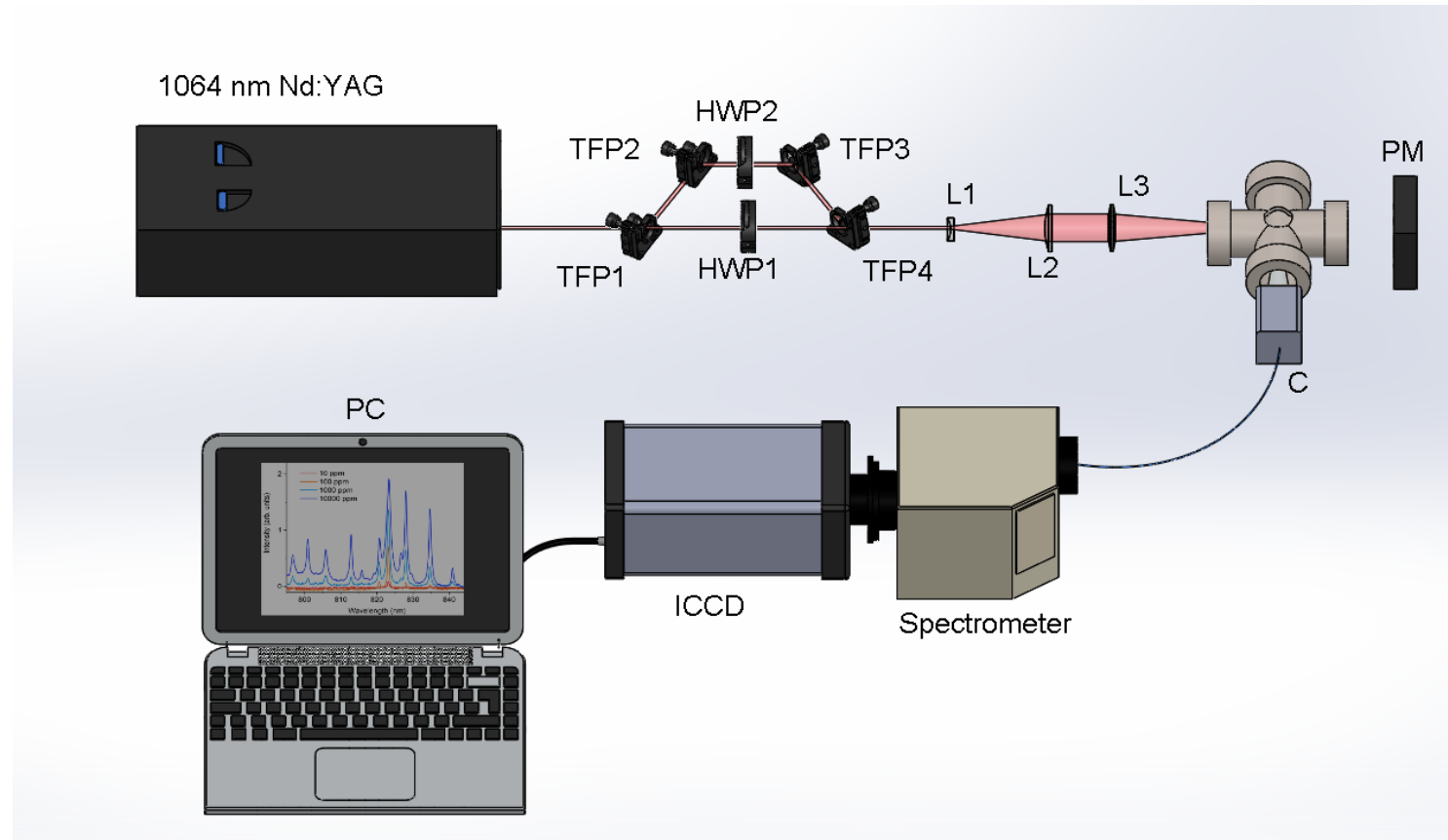


Cremers et al. *Appl. Spectrosc.*, **38**, 721-729 (1984).



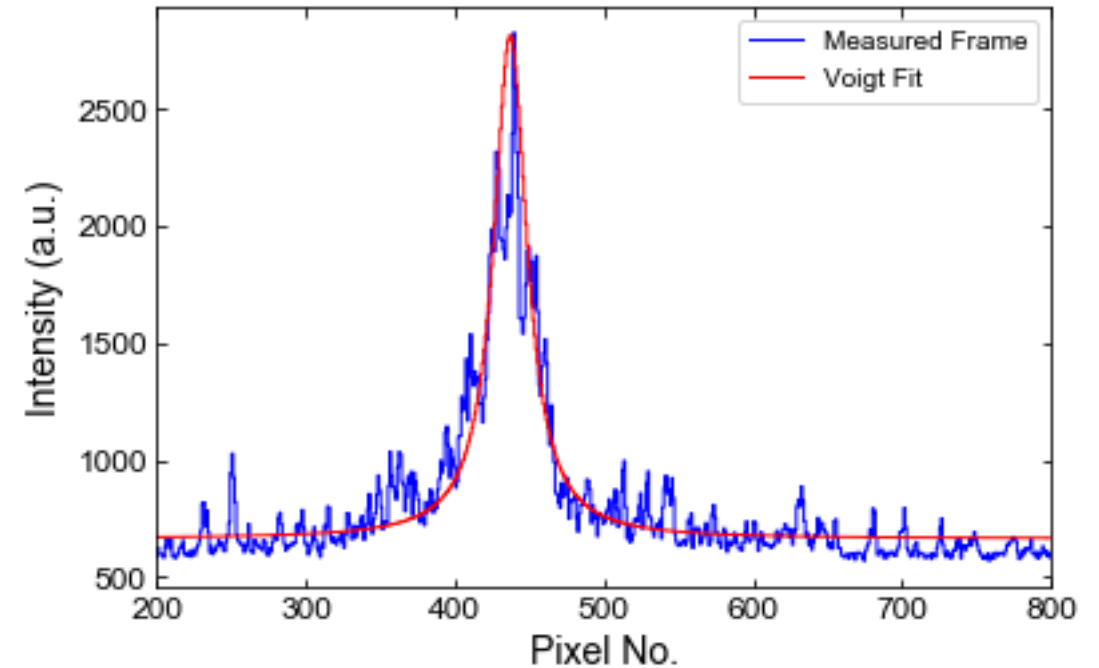
Windom et al. *Spectrochim. Acta Part B At. Spectrosc.*, **61**, 788-796 (2006).

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



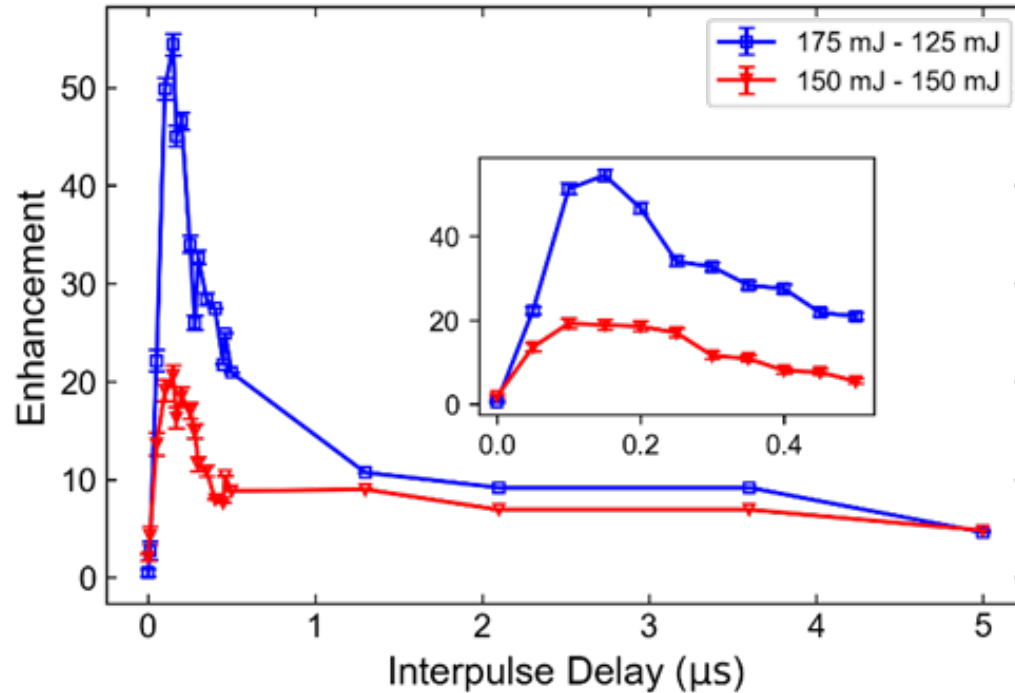
An analytical framework for spectral analysis is used to quantify the enhancement

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

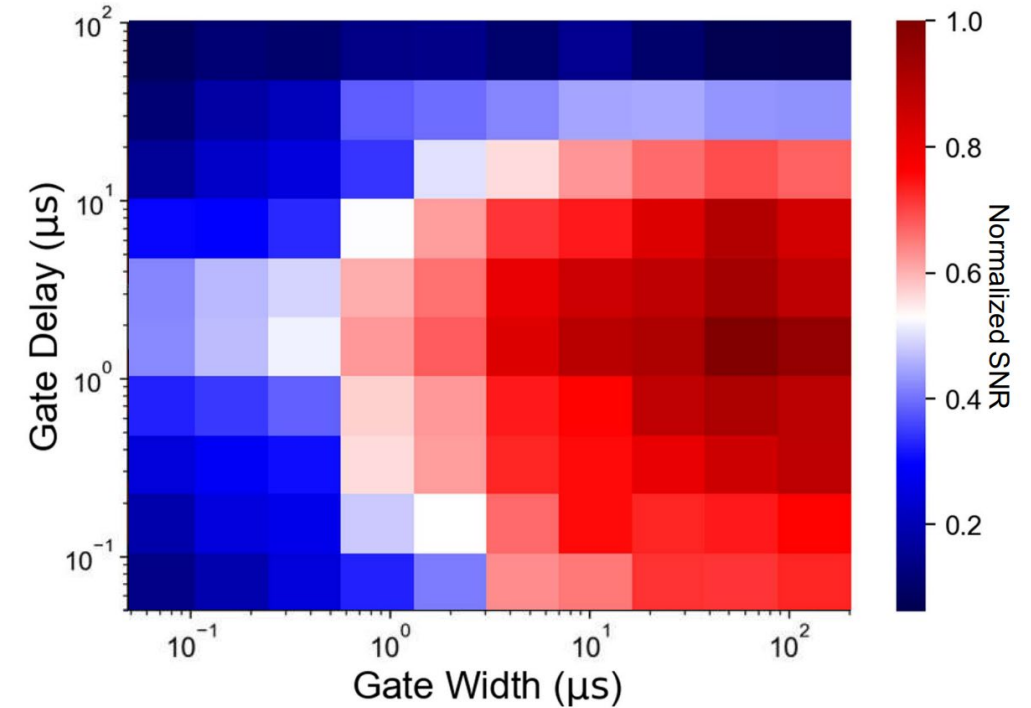


The large parameter space requires several optimizations

Enhancement vs. Interpulse Delay

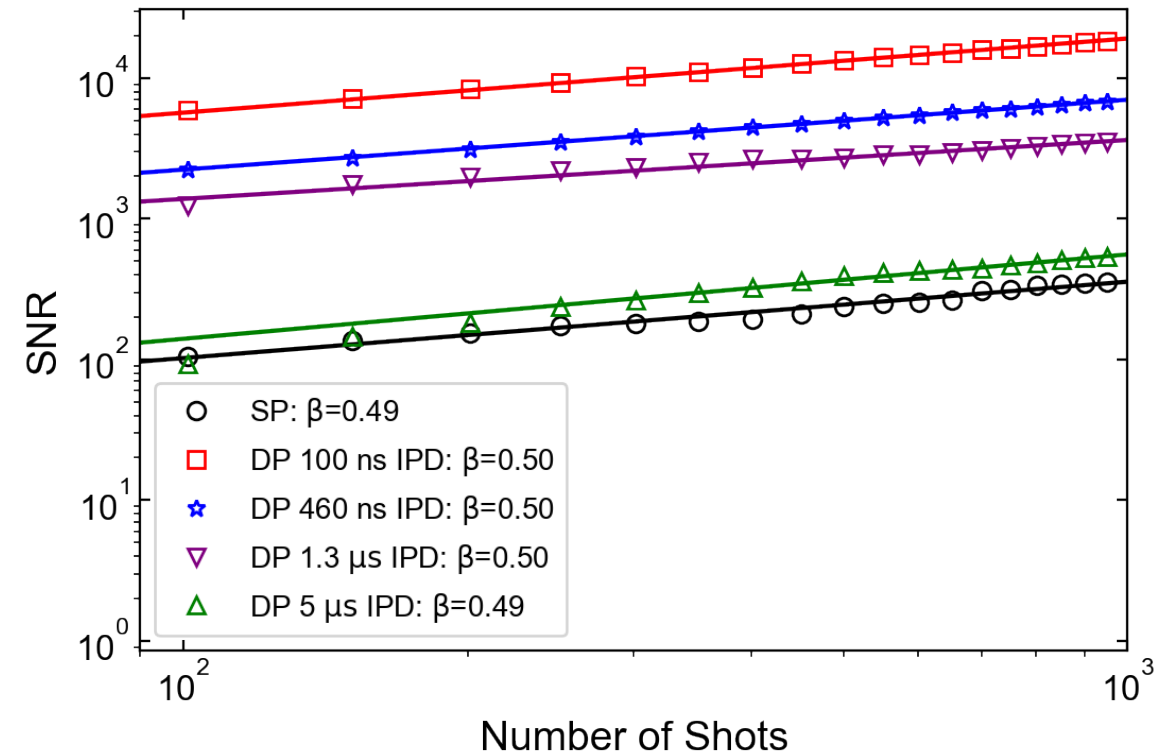
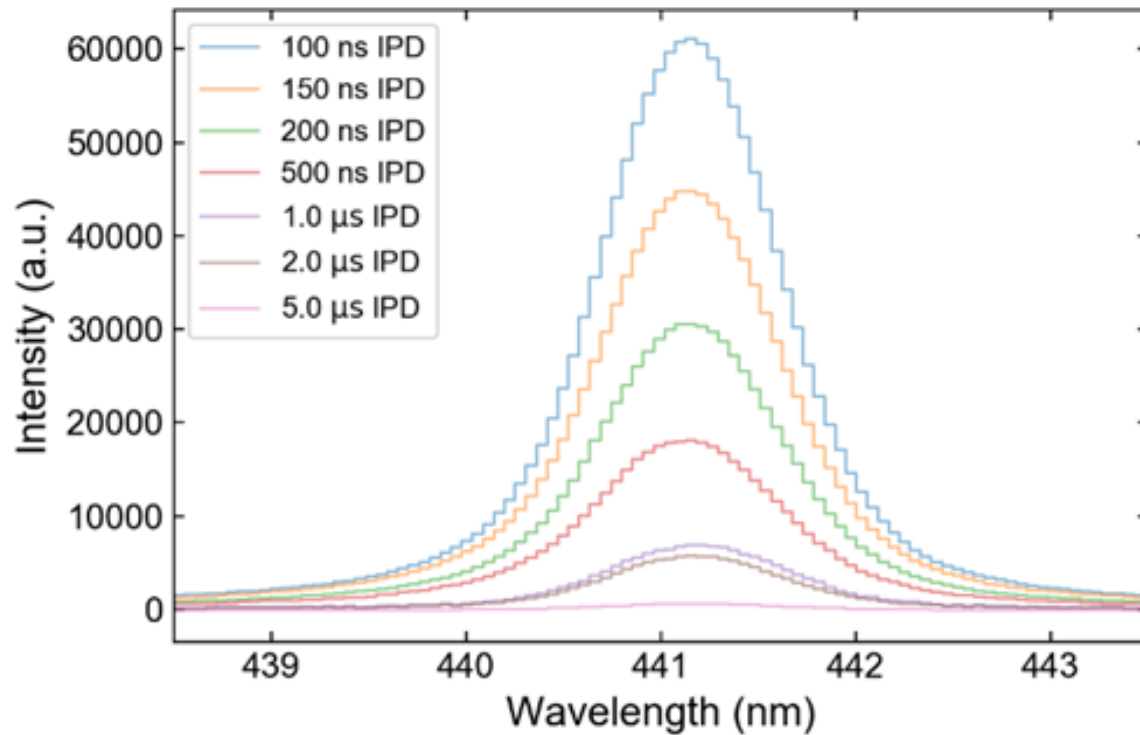


SNR vs. ICCD Recording Parameter



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

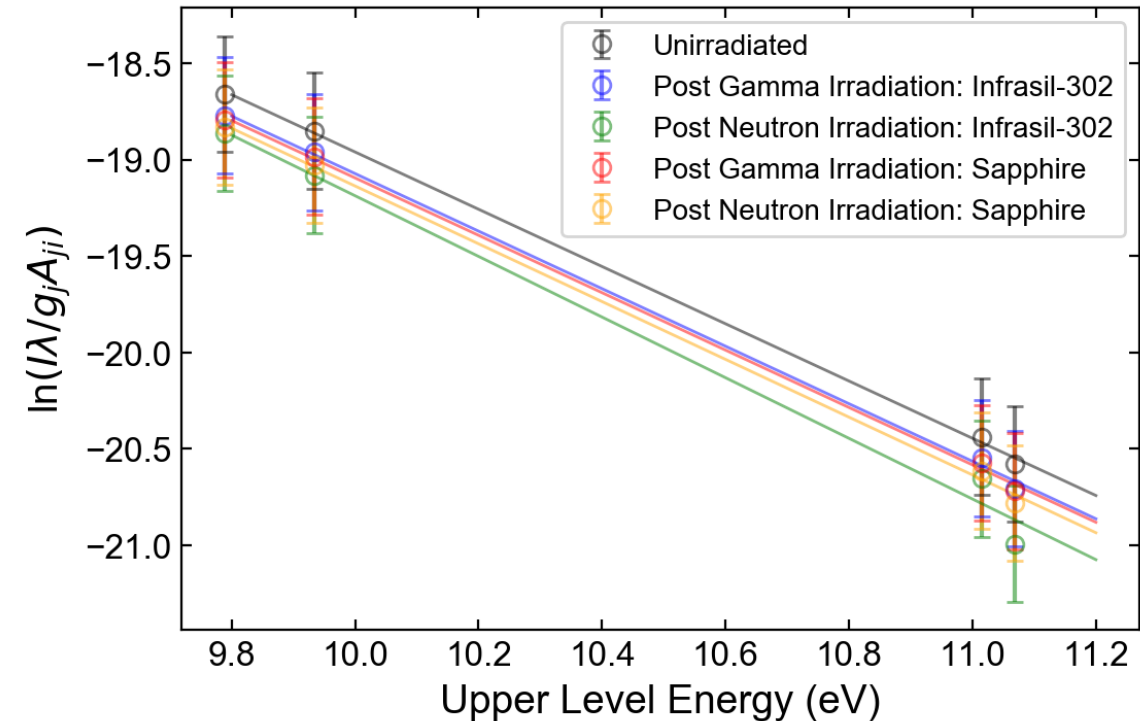
A ~50x enhancement is observed in the 1 ppm signal at optimal conditions



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
 - Rarefaction of the gaseous environment

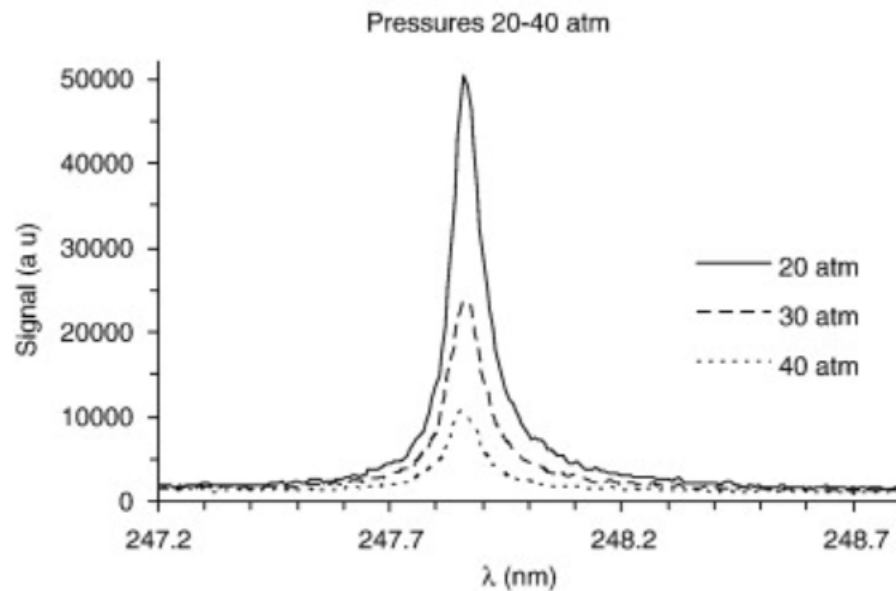


Burger et al., *J. Anal. At. Spectrom.* **36**, 824-828 (2021).

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 850° C and 13 MPa

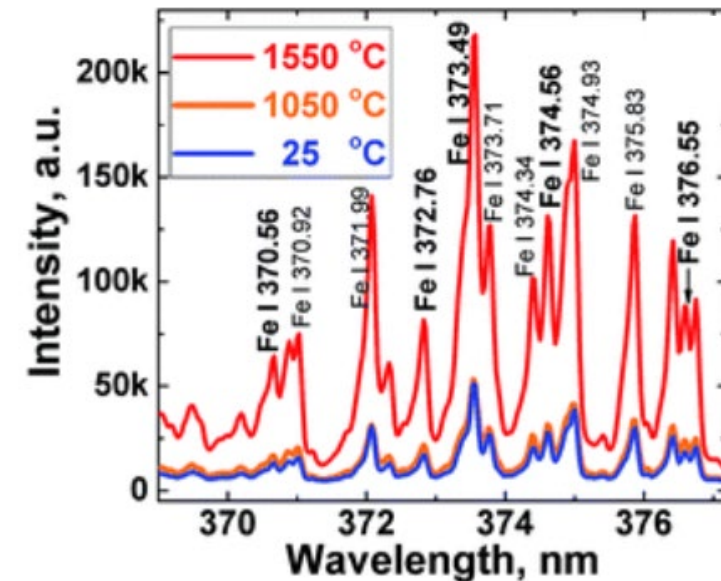


Increased Pressure



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)

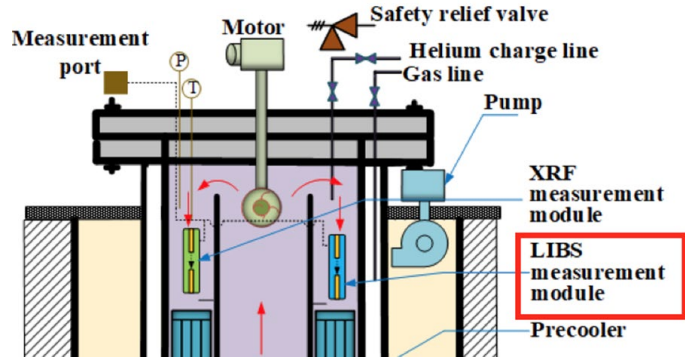
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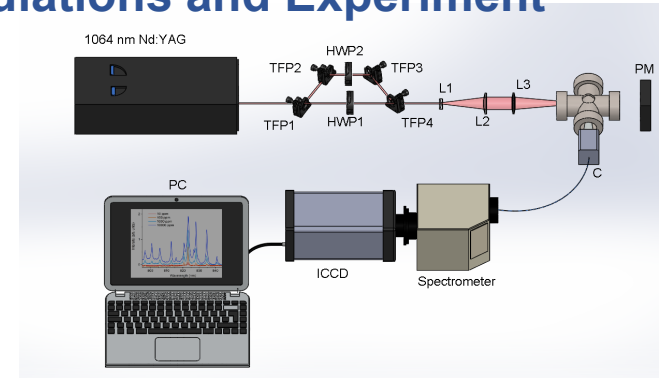
This research was performed under appointment to the Nuclear Nonproliferation International Safeguards Fellowship Program sponsored by the Department of Energy, National Nuclear Security Administration's Office of International Nuclear Safeguards (NA-241)

Summary

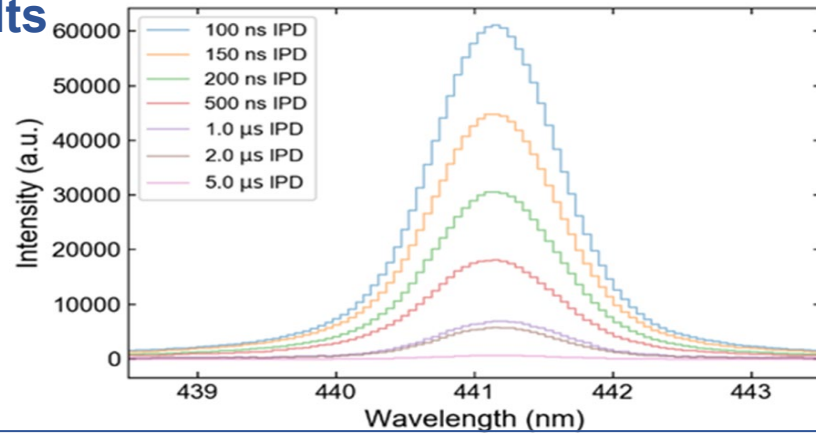
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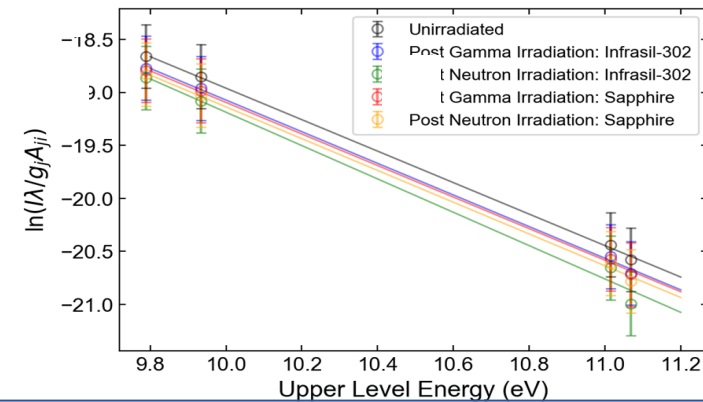
Calculations and Experiment



Results



Future Directions



Questions?