Irradiation of Optical Components of *In-Situ* Laser Spectroscopic Sensors

Advanced Sensors and Instrumentation (ASI) Annual Program Webinar
October 30 – November 2, 2023

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Project Overview (FY 2020–2023)

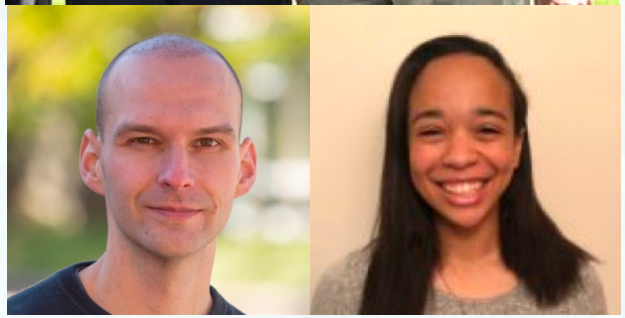
Goal and Objective: understand the effect of radiation damage on the performance of optical spectroscopic sensors with special emphasis on:
(1) nonlinear refractive index
(2) transient radiation-induced absorption
(3) concurrent radiation damage and thermal annealing

Schedule:

- Year 1: Procure samples; develop mobile PIE system
- Year 2: Evaluate neutron activation; construct and test heating setup; conduct gamma irradiation with post-heating
- Year 3: Conduct neutron irradiation with post-heating
- Year 4: Conduct gamma and neutron irradiation with concurrent heating

The project period ended on September 30, 2023.

Research Team and Collaborations



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Paul Marotta (MicroNuclear)

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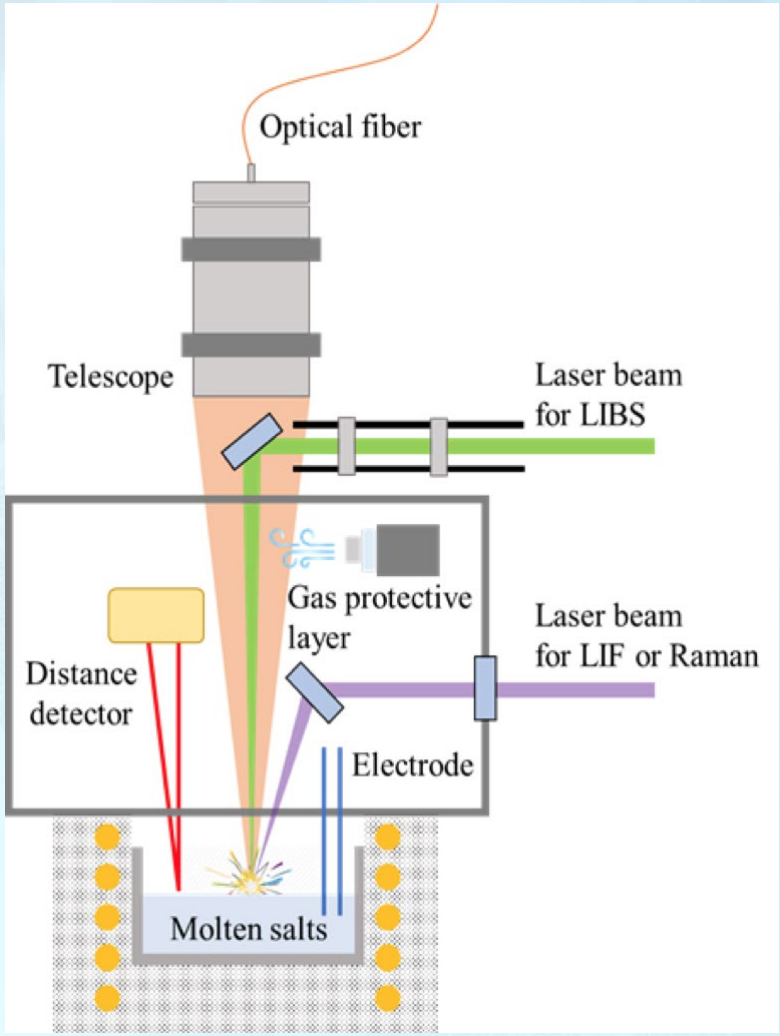
Sungyeol Choi (Seoul National University – INERI collaborator)

Christian Petrie (ORNL – collaborator)

Sylvain Gerard (Université Jean Morret – Saint-Étienne - collaborator)

Technology Impact

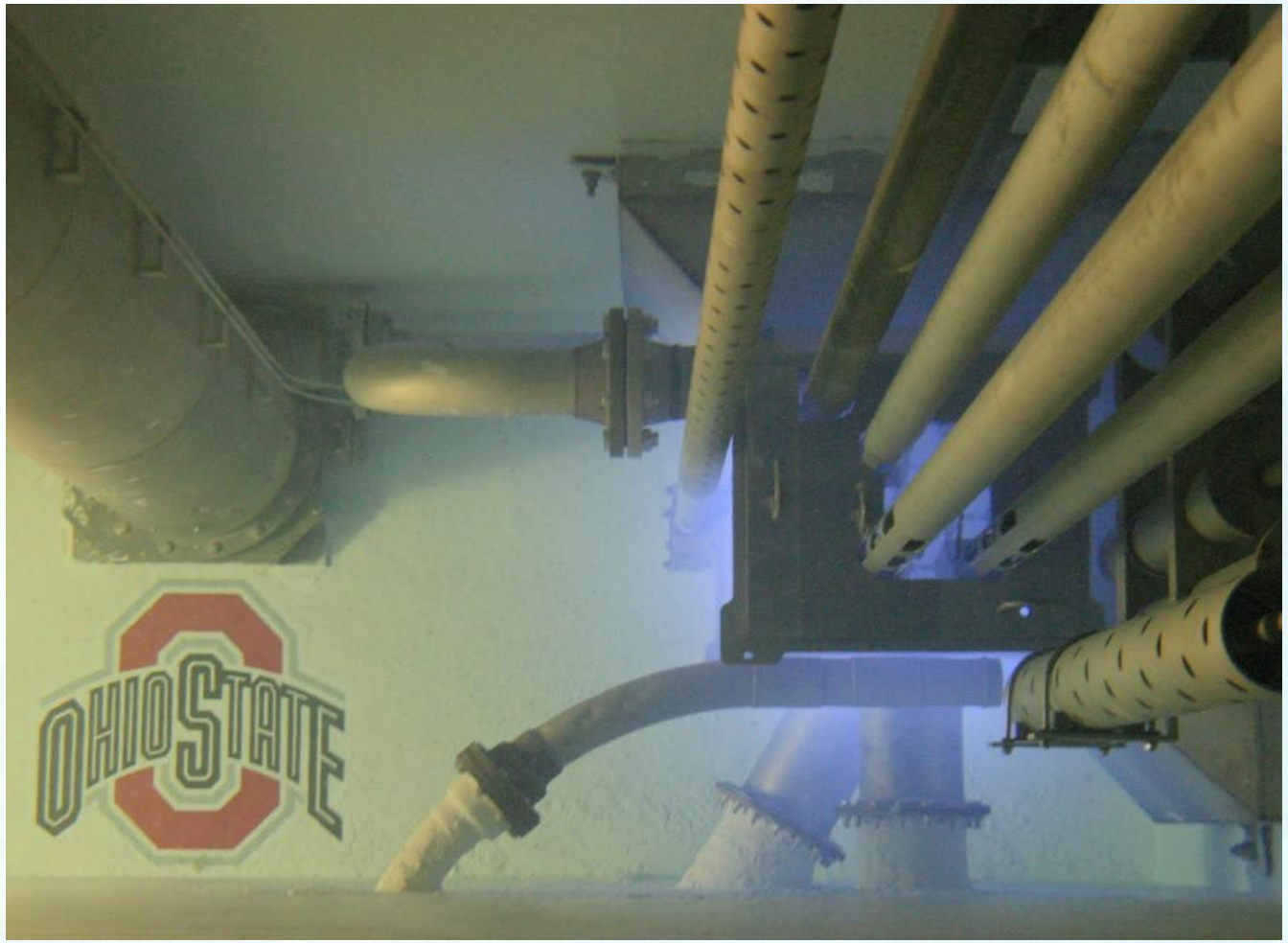
- Develop an improved understanding of radiation damage in optical materials in conditions relevant for their operation in real-time optical sensors
- First-ever attempt to quantify the effect of irradiation on nonlinear optical properties of materials
- Real-time, *in-situ* measurements of important operational parameters in advanced nuclear systems → **safety + economic performance**
- **Cross-cutting impact: design and concept of operation for a wide range of optical instrumentation in nuclear applications**



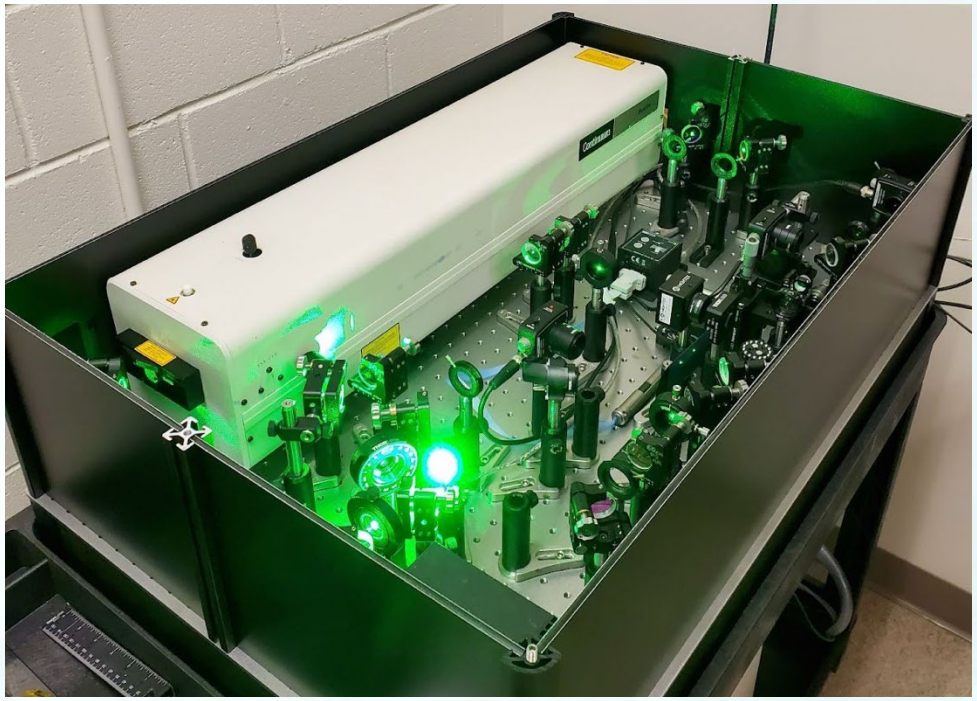
LIBS of molten salts

Irradiation Facilities

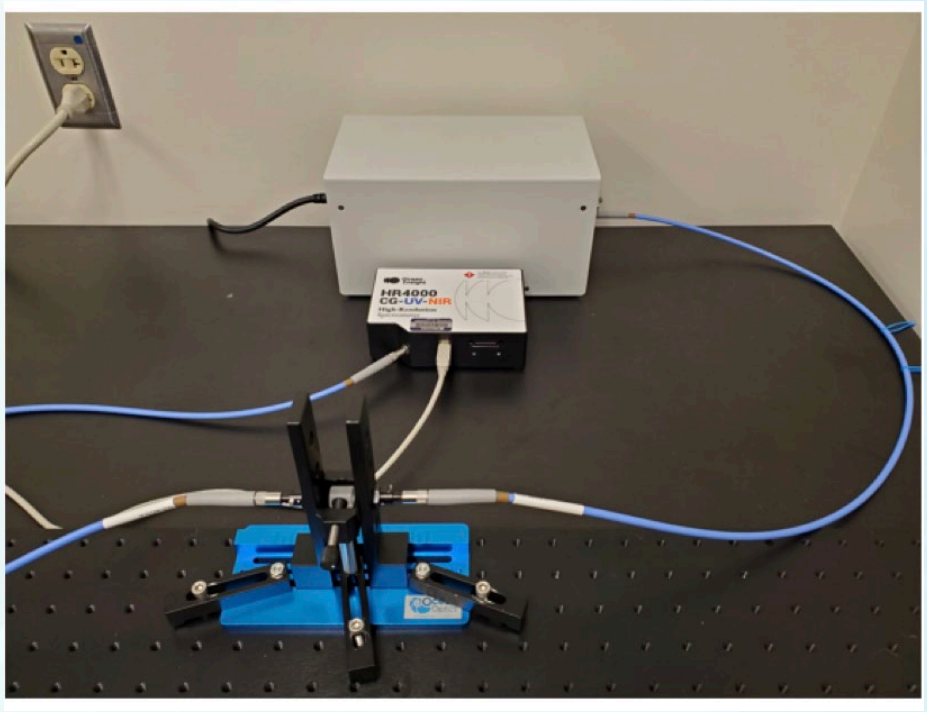
- Sample irradiation and thermal annealing at the OSU Nuclear Reactor Laboratory (DOE NSUF)
- Gamma irradiation at the PSU Radiation Science & Engineering Center (DTRA IIRM-URA)



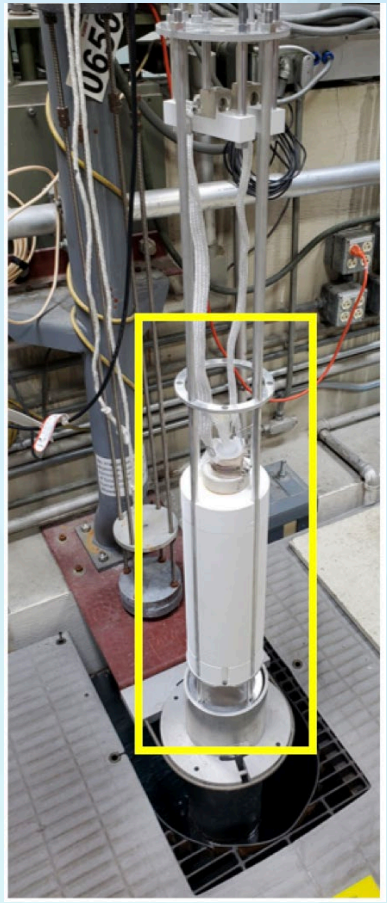
Experimental Setup



PIE: Z-scan



PIE: absorption

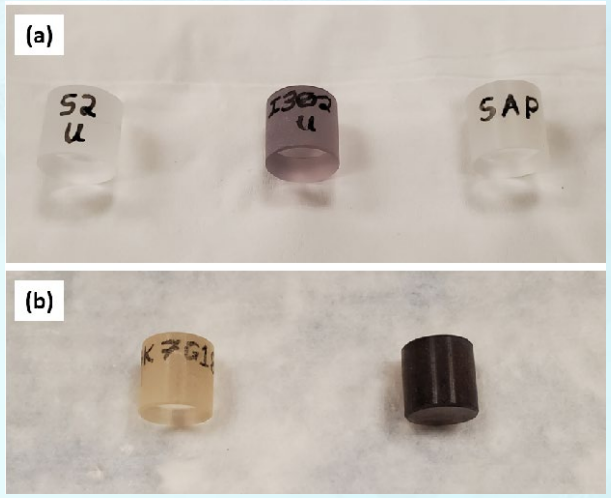


Sample furnace

Irradiation Conditions and Samples

Source	Dose/Fluence	Anneal Type	Temp.	Time
^{60}Co	600 krad 1.2 Mrad 3.4 Mrad	Post	200 °C 400 °C 600 °C 800 °C	30 min. 30 min. 30 min. 30 min.
^{60}Co	600 krad 1.2 Mrad 3.4 Mrad	Concurrent	800 °C	Duration
Reactor	3.4×10^{16} n · cm^{-2} (42 Mrad) 1.7×10^{17} n · cm^{-2} (211 Mrad)	Post	200 °C 400 °C 600 °C 800 °C	30 min. 30 min. 30 min. 30 min.
Reactor	3.4×10^{16} n · cm^{-2} (42 Mrad) 1.7×10^{17} n · cm^{-2} (211 Mrad)	Concurrent	800 °C	Duration

Material	Vendor	Type	OH Content
High-OH Fused Silica	Heraeus	Spectrosil 2000	$\leq$ 1300 ppm
Low-OH Fused Silica	Heraeus	Infrasil 302	$\leq$ 8 ppm
Sapphire	Guild Optical Associates	Optical Grade	



Nonlinear Refractive Index and Absorption

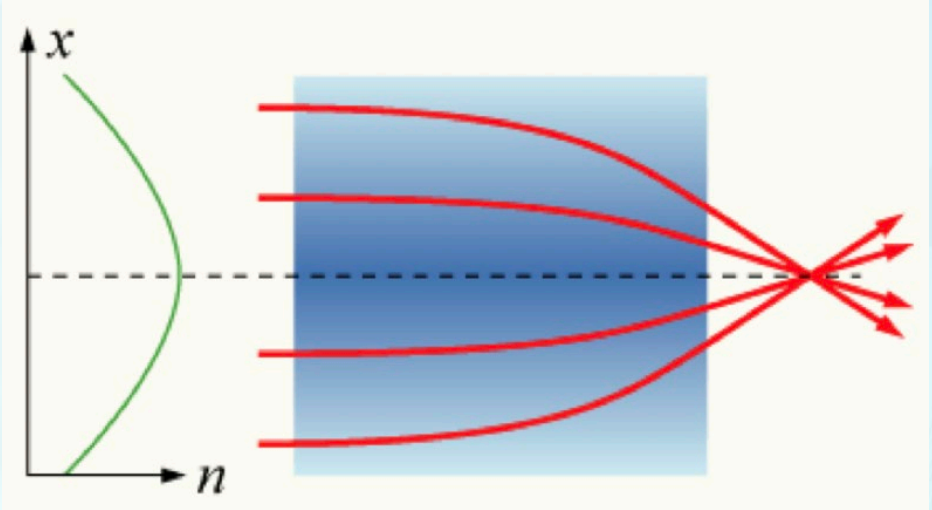
Materials exhibit nonlinear optical properties caused by variation of induced electronic polarization (P) with the applied electric field (E)

$$P = \epsilon_0 \chi^{(1)} E + \epsilon_0 \chi^{(2)} E^2 + \boxed{\epsilon_0 \chi^{(3)} E^3} + \ldots$$

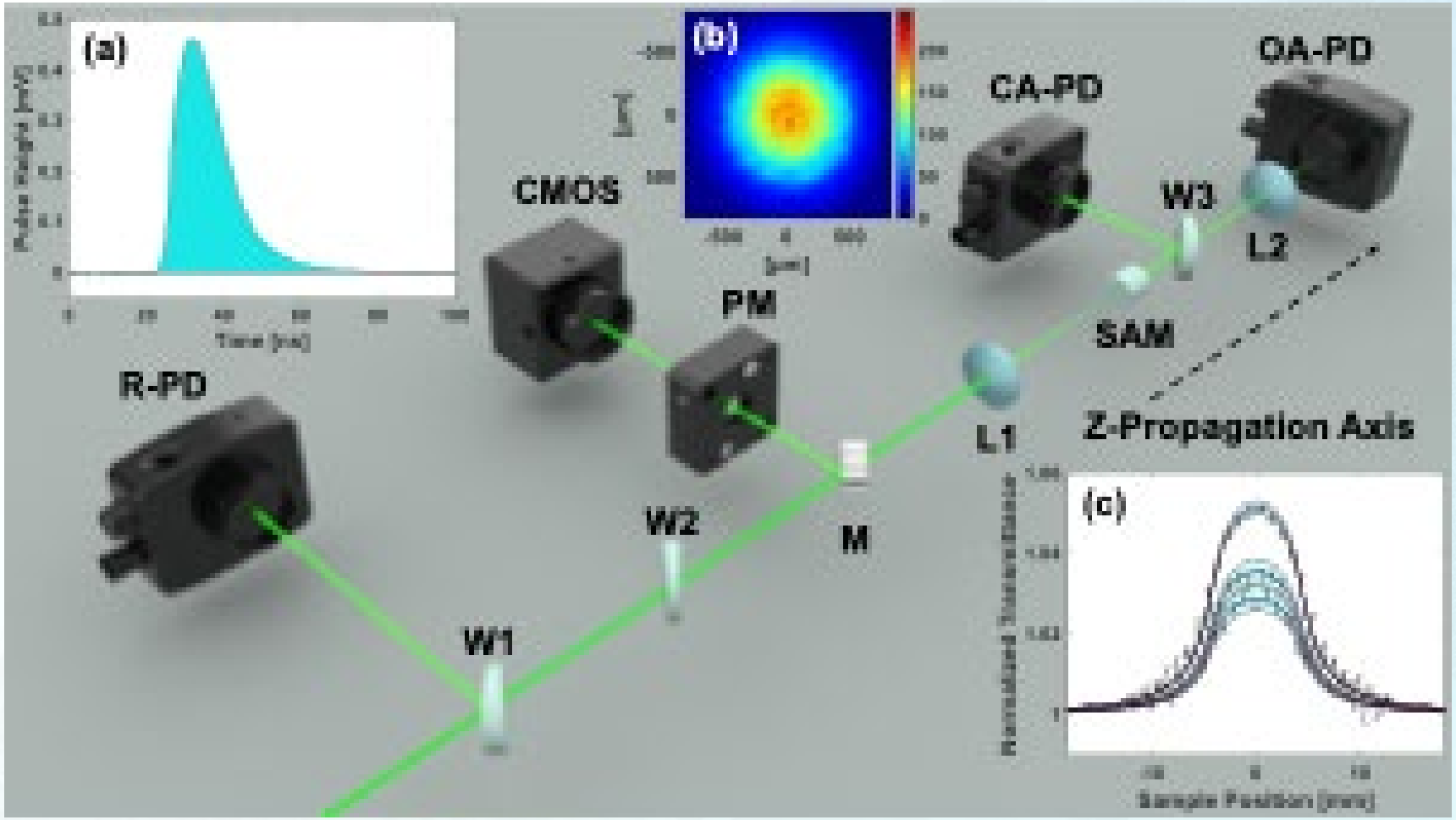
The third-order nonlinear susceptibility leads to processes such as third-harmonic generation, two-photon absorption, and the intensity-dependent refractive index.

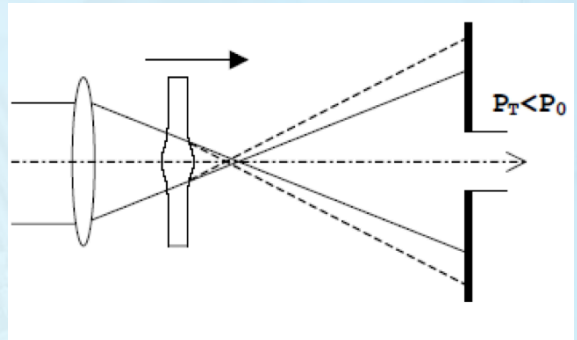
$$n = n_0 + n_2 I \left\{ \begin{array}{l} \text{self-focusing} \\ \text{nonlinear absorption} \end{array} \right.$$

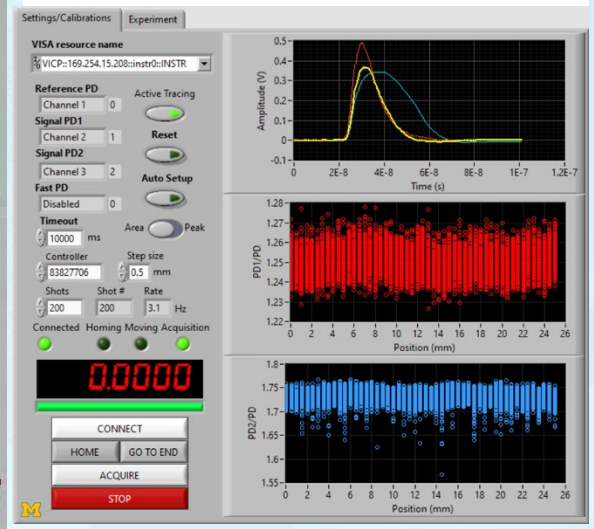
$$n_2 = \frac{12\pi^2 \chi^{(3)}}{n_0^2}$$



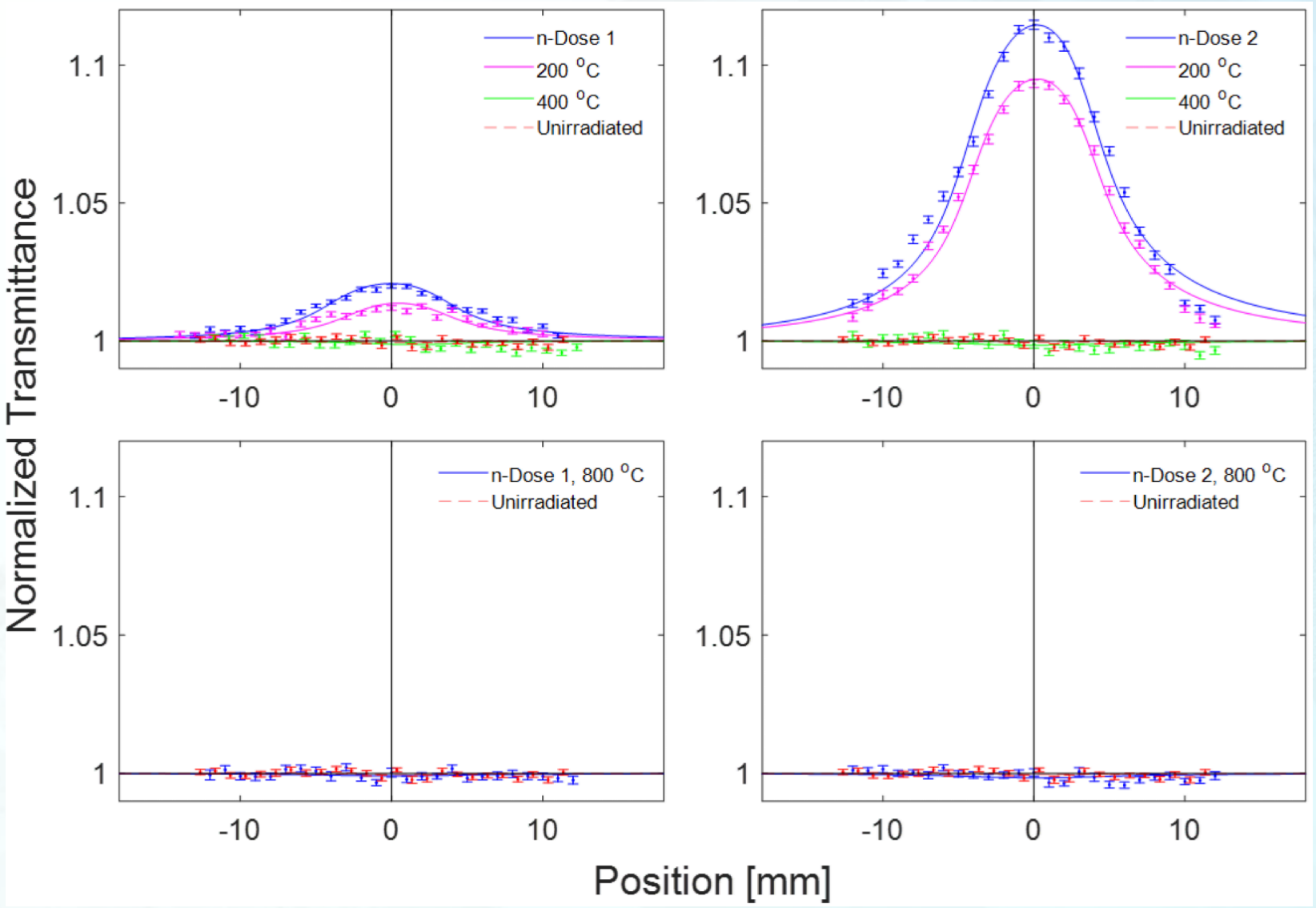
Measurement of Nonlinearities: Z-scan







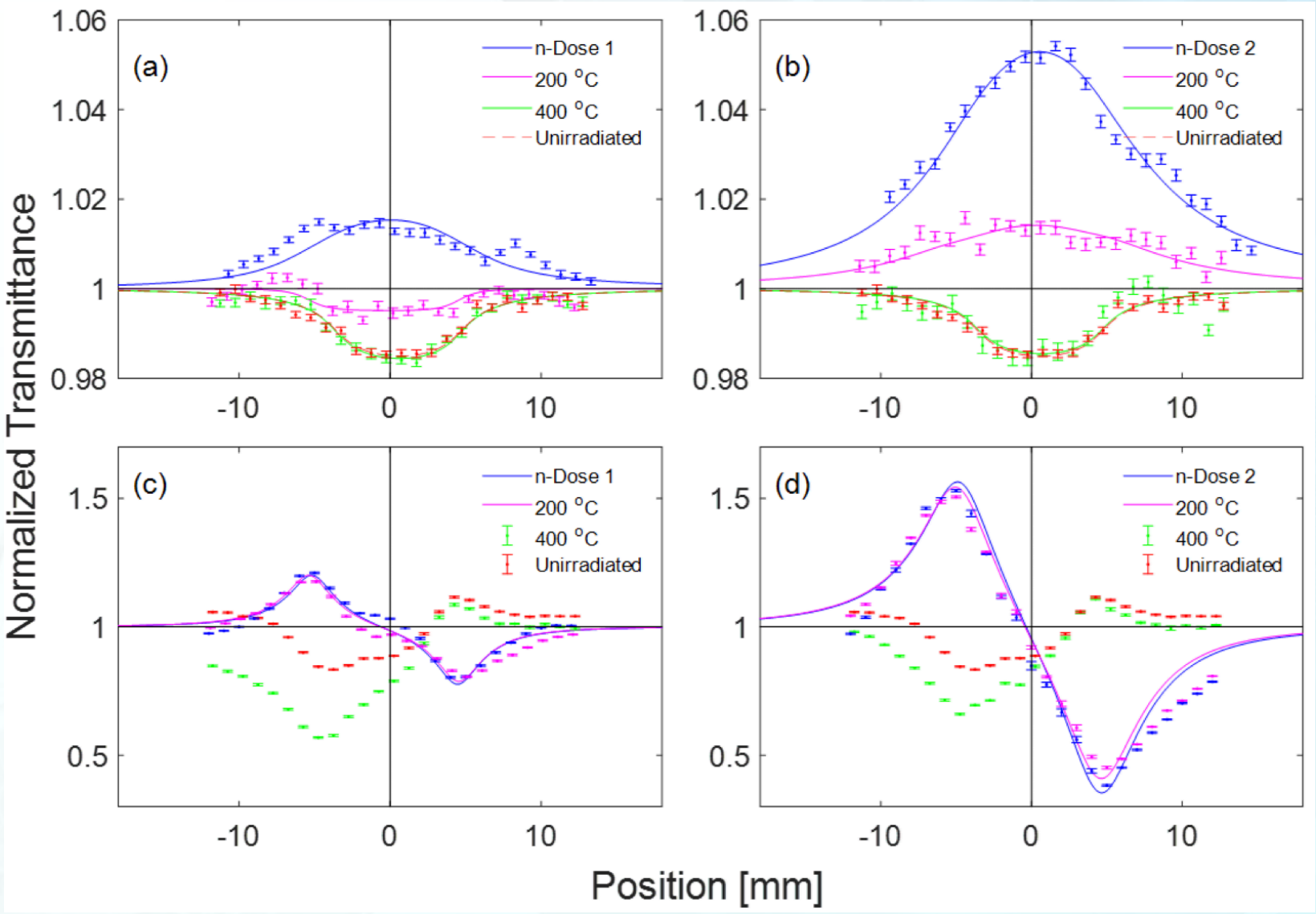
Negative Nonlinear Absorption in Neutron-Irradiated Sapphire



Sapphire

Annealing

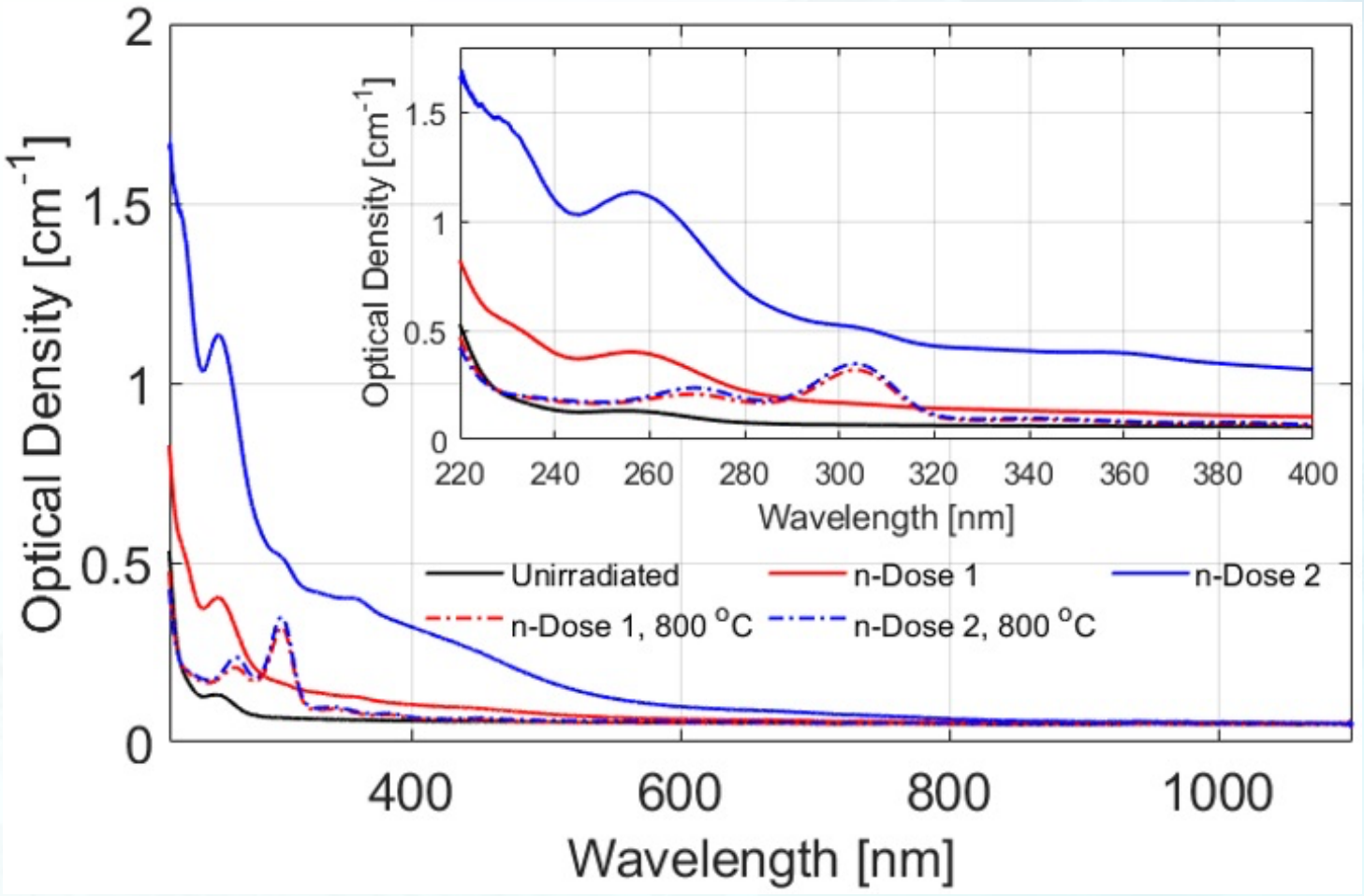
BK7G18 Neutron Irradiation: Nonlinear Absorption and Refraction



Nonlinear absorption

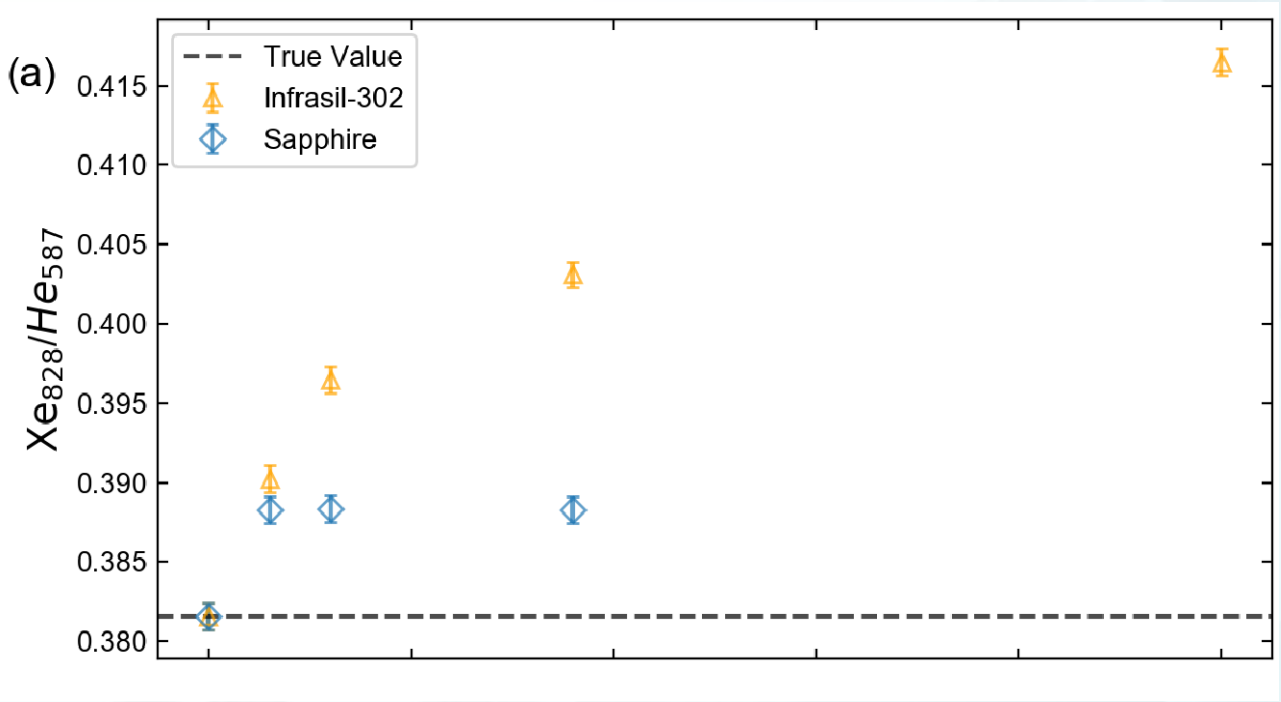
Nonlinear refraction

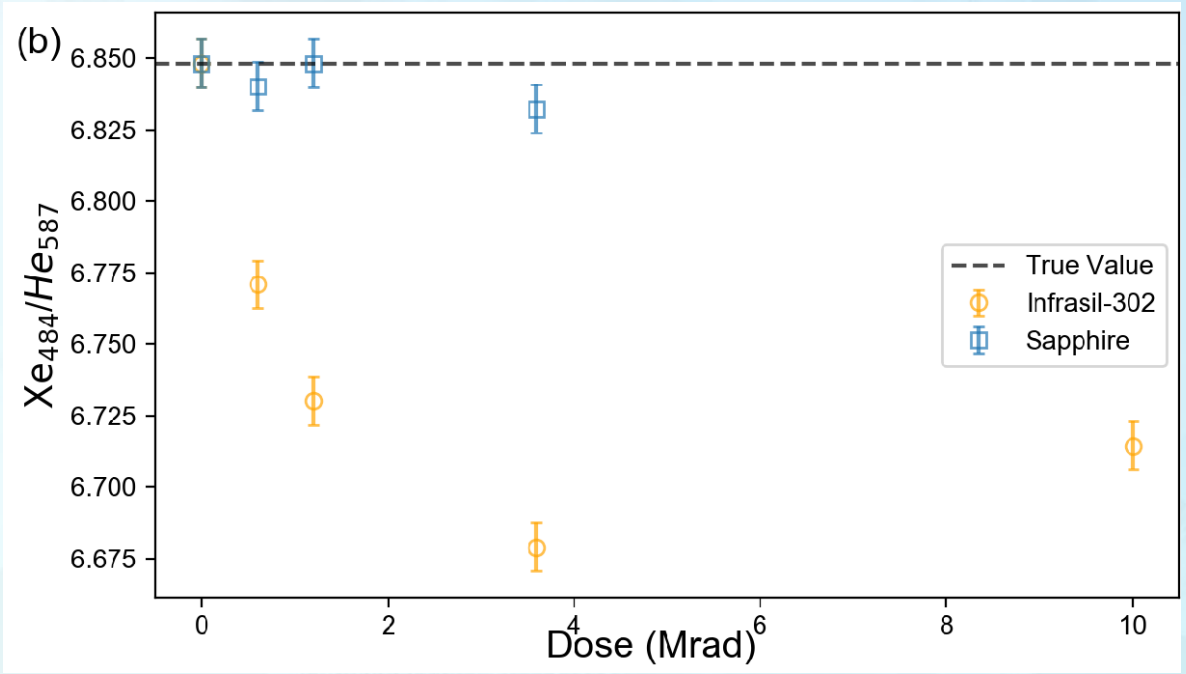
RIA in Sapphire with Concurrent Annealing



n-Dose 1: 3.4×10^{16} n · cm^{-2}
n-Dose 2: 1.7×10^{17} n · cm^{-2}

Effect of Irradiation on LIBS Instrumentation

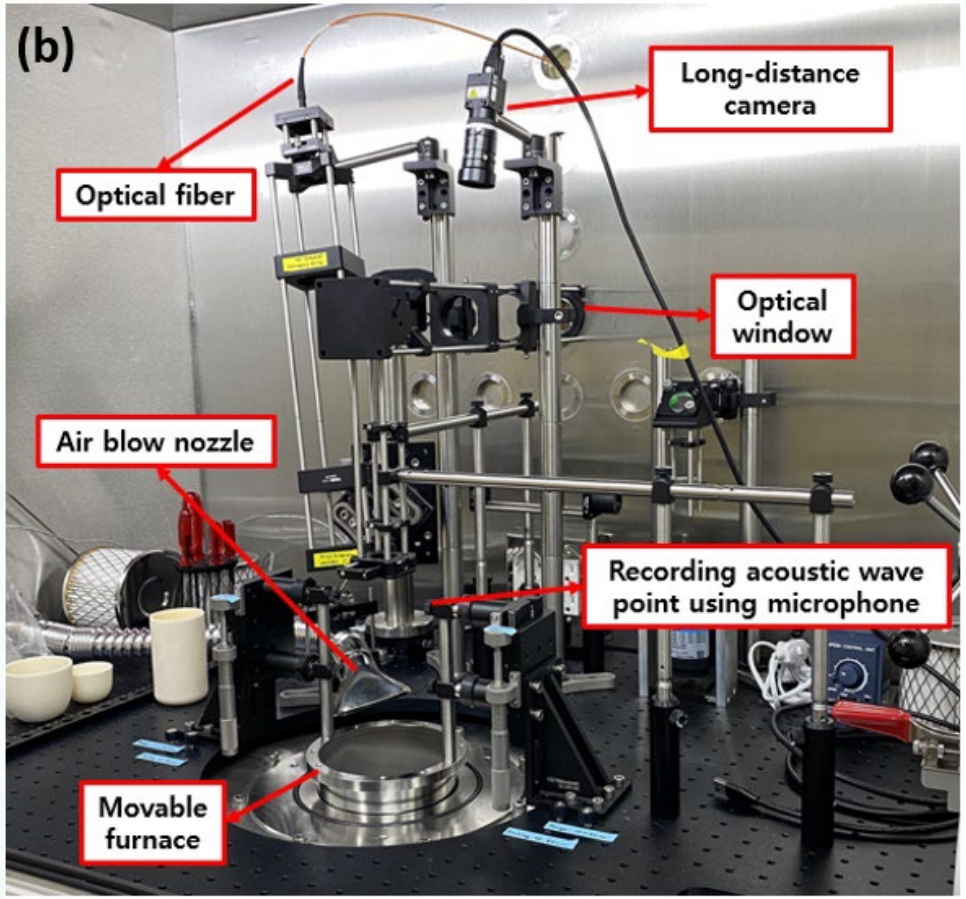


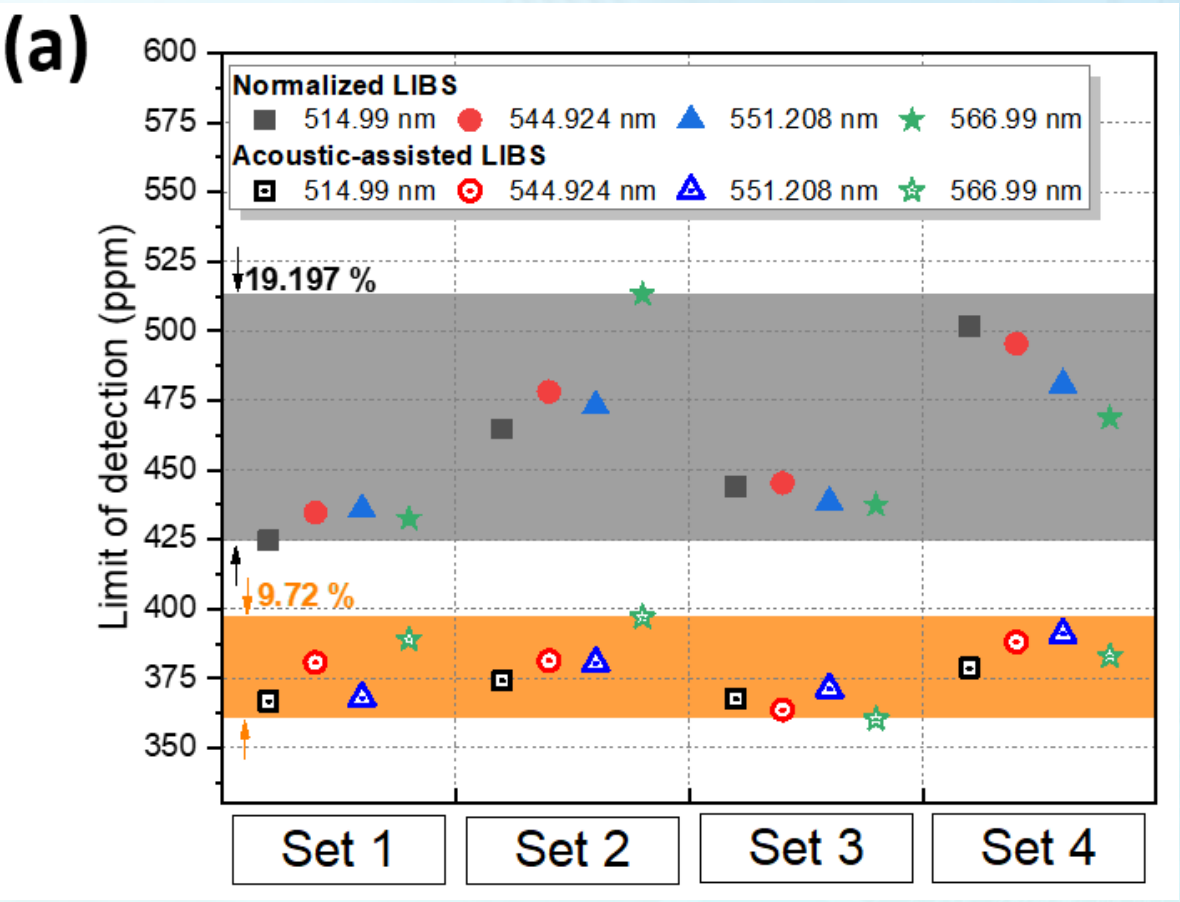


- Relative line intensities are used to determine relative component concentrations
- Irradiation can lead to systematic errors and calculation of plasma parameters used in normalization and correction for self-absorption.

INERI Collaboration (Seoul National University)

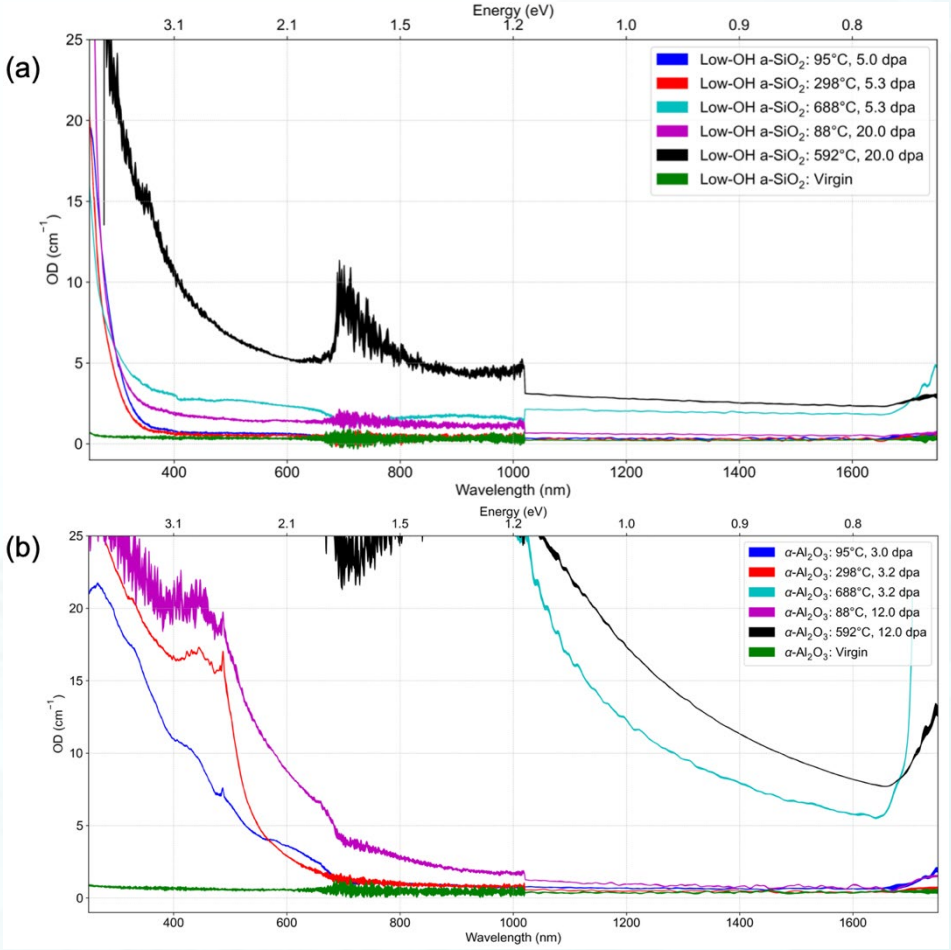
- Detection of corrosion products (Ni, Fe, Cr) in molten salts
- Cerium chloride (surrogate for plutonium) in LiCl-KCl eutectic
- Acoustic normalization





RTE Project (Collaboration with ORNL)

Radiation-induced Attenuation and Nonlinear Optical Properties of Fused Silica and Single-crystal Sapphire



- RIA for α-Al_2O_3 and a-Al_2O_3 in samples irradiated to high neutron fluence (~10^{22} n/cm^2) at high temperatures
- No significant change in low-OH a-SiO_2 samples irradiated to a higher dose
- Low-OH samples that were irradiated to a higher showed increased absorption at shorter wavelengths, compared to the lower-dose samples
- Sapphire OD spectra largely resemble those obtained in our previous measurements, but with an overall increase in magnitude

Journal Papers

- B. W. Morgan, M. P. Van Zile, C. M. Petrie, P. Sabharwall, M. Burger, and I. Jovanovic, “Radiation-Induced Negative Nonlinearities in Fused Silica, Sapphire, and Borosilicate Glass,” *Journal of Nuclear Materials* 582, 154486 (2023).
- L. Garrett, B. W. Morgan, M. Burger, Y. Lee, H. Kim, S. Choi, P. Sabharwall, and I. Jovanovic, “Impact of Glass Irradiation on Laser-Induced Breakdown Spectroscopy Data Analysis,” *Sensors* 23, 691 (2023).
- B. W. Morgan, M. P. Van Zile, C. M. Petrie, P. Sabharwall, M. Burger, and I. Jovanovic, “Optical Absorption of Fused Silica and Sapphire Exposed to Neutron and Gamma Radiation with Simultaneous Thermal Annealing,” *Journal of Nuclear Materials* 570, 153945 (2022).
- Y. Lee, S. Yoon, H. Kim, N. Kim, W. Yang, D. Kang, M. Burger, I. Jovanovic, and S. Choi, “In-situ measurement of Ce concentration in high-temperature molten salts using acoustic-assisted laser-induced breakdown spectroscopy with gas protective layer,” *Nuclear Engineering and Technology (2022)* https://doi.org/10.1016/j.net.2022.07.014.
- B. W. Morgan, M. Van Zile, P. Sabharwall, M. Burger, and I. Jovanovic, “Gamma-radiation-induced negative nonlinear absorption in quartz glass,” *Optical Materials Express* 12, 1188-1197 (2022).
- B. W. Morgan, M. Van Zile, P. Sabharwall, M. Burger, and I. Jovanovic, “Post-Irradiation Examination of Optical Components for Advanced Fission Reactor Instrumentation,” *Review of Scientific Instruments* 92, 105107 (2021).

Published this year Published earlier during this project

Conference Presentations

- B. W. Morgan, M. Van Zile, P. Sabharwall, M. Burger, and I. Jovanovic, “Linear and Non-linear Optical Characterization of Glass and Sapphire for Optical Instrumentation of Advanced Fission Reactors,” ANS Winter Meeting, Washington, DC, November 12–15, 2023.
- L. J. Garrett, B. Morgan, M. Burger, Y. Lee, H. Kim, S. Choi, and I. Jovanovic, “Impact of Glass Radiation Damage on Optical Spectroscopy,” ANS Winter Conference, Phoenix, AZ, November 13–17, 2022.
- B. Morgan, M. Burger, and I. Jovanovic, “Linear and Nonlinear Optical Properties of Fused Silica and Sapphire in Extreme Radiation and Thermal Environments,” IEEE Nuclear & Space Radiation Effects Conference, Provo, UT July 18–22, 2022.
- B. W. Morgan, M. Van Zile, P. Sabharwall, M. Burger, and I. Jovanovic, “Radiation-induced Negative Nonlinear Absorption in Glass and Sapphire,” Conference on Lasers and Electro-Optics, San Jose, CA, May 15-20, 2022.
- B. Morgan, M. Van Zile, P. Skrodzki, X. Xiao, P. Sabharwall, P. Marotta, M. Burger, and I. Jovanovic, “Post-Irradiation Examination of Irradiated Optical Components of In-Situ Spectroscopic Sensors for Advanced Fission Reactors,” ANS Winter Meeting, November 30–December 3, 2021.
- B. Morgan, P. Skrodzki, M. Burger, P. Sabharwall, P. Marotta, and I. Jovanovic, “Post-Irradiation Examination System Development for Irradiated Optical Components of In-Situ Spectroscopic Sensors,” ANS Winter Conference [online], November 15-19, 2020.

Summary of Accomplishments

- Mobile PIE system constructed and validated
- PIE system moved and operated at OSU NRL
- Constructed and operated thermal annealing furnaces
- Neutron/gamma irradiations completed; final analysis pending
- Established collaborations with ORNL, SNU, and UJM-SE
- RTE project with ORNL
- INERI collaborative project with SNU
- 6 papers published; 6 conference presentations; 1 dissertation

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Key Research Findings

- Negative nonlinear absorption observed for the first time in bulk optical materials that include fused silica, sapphire, and BK7 crown glass
- Negative nonlinear refraction observed for the first time in BK7G18
- Additional information obtained on the ability of thermal annealing to repair radiation damage *in situ*
- RTE: differences between high-OH and low-OH content glass at high radiation doses
- Effect on LIBS instrumentation quantified

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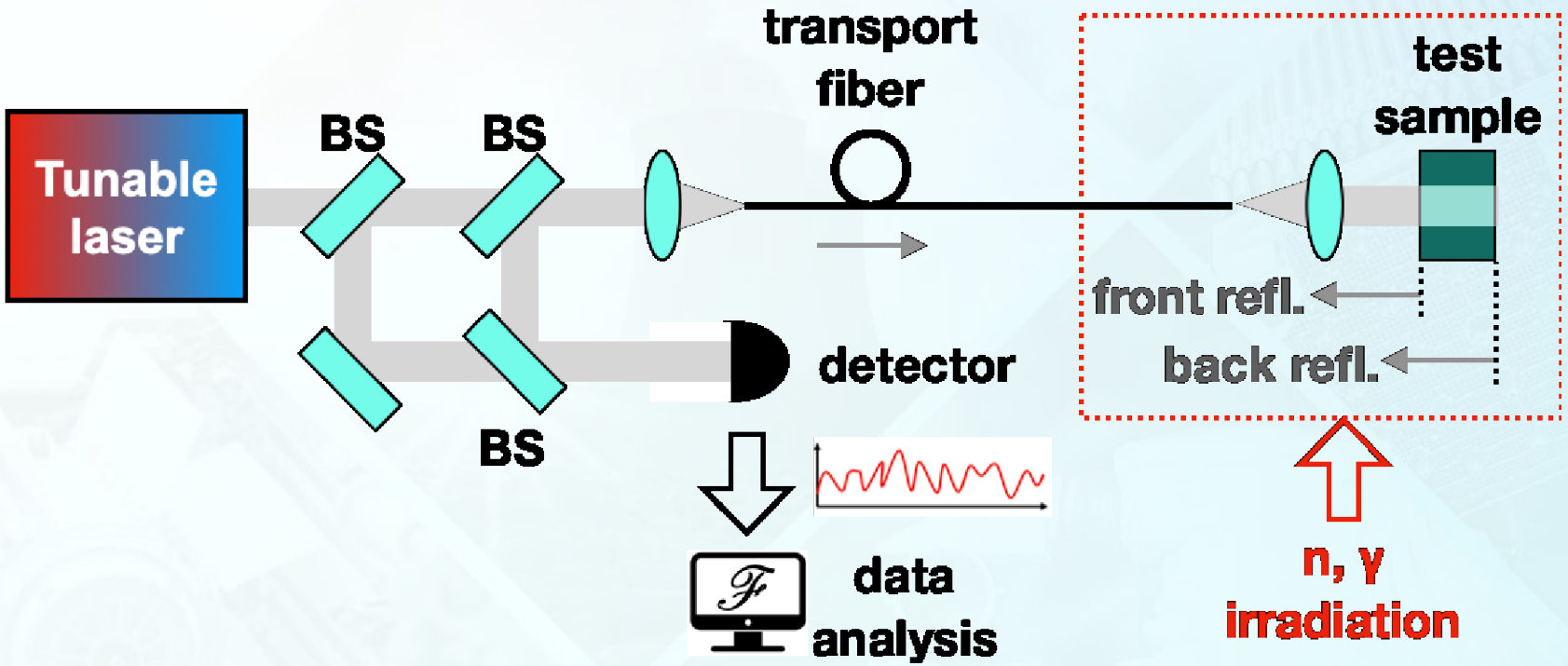
Future Work

- Time-resolved measurement of saturable nonlinear absorption
- **True *in-situ*, real-time** RIA measurement using frequency-domain reflectometry



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Thank You

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