Optical measurements of irradiated crystals for PALEOCCENE

Spectroscopy measurements of neutron irradiated crystals

Xianyi Zhang (LLNL), Felicia Sutanto (LLNL), Stuti Surani (PSU)

Mineral Detection of Neutrinos and Dark Matter
01/11/2023

LLNL-PRES-858984

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344. Lawrence Livermore National Security, LLC

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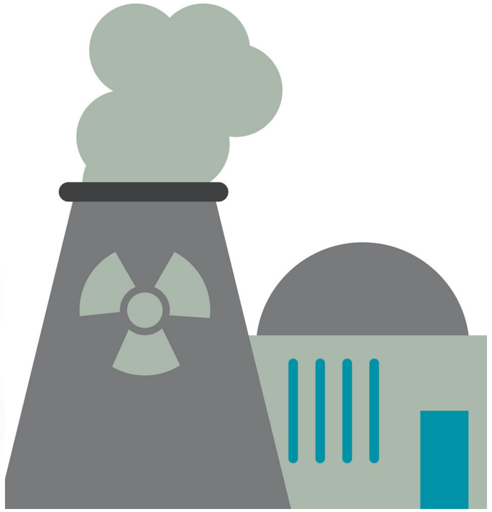
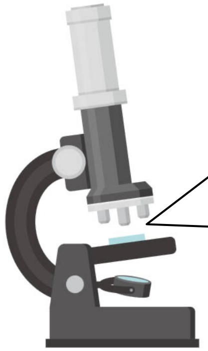
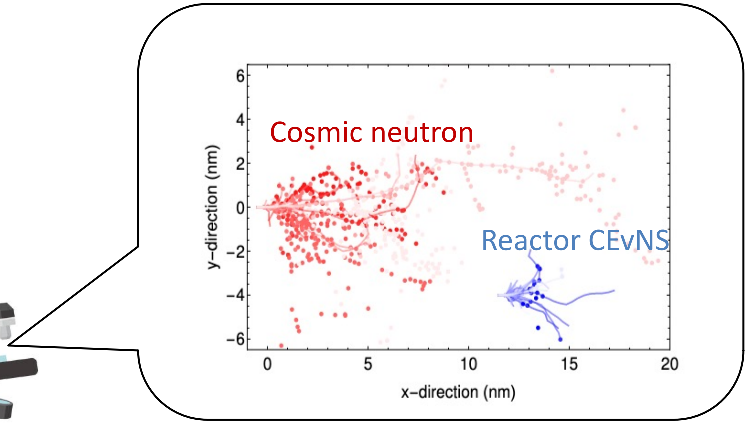
Ultimate goals of the LLNL work

- Measure CEvNS from reactor neutrinos for nuclear non-proliferation application
- Passive detection – very attractive to application fields
- Aim to measure nuclear recoils to the keV scale with color centers
- Requires efficient counting and energy reconstruction



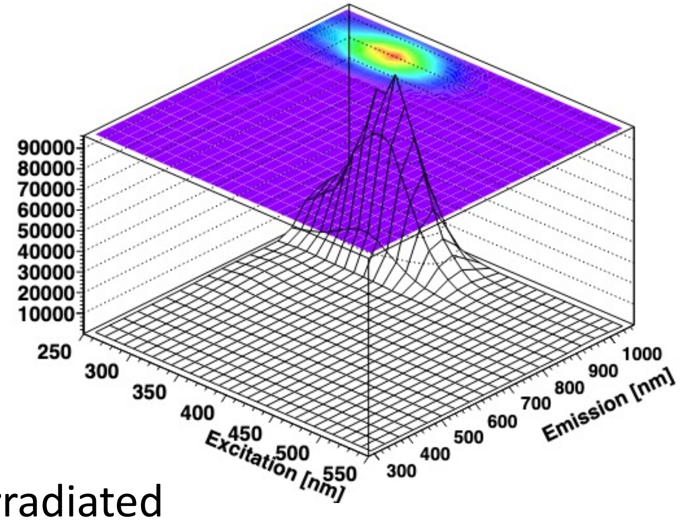




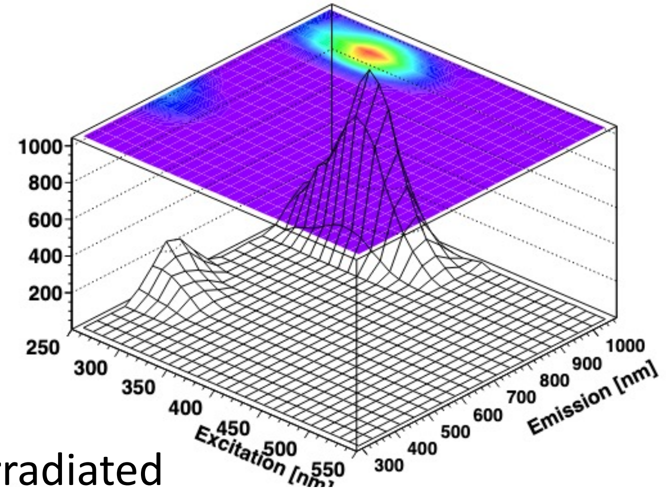
Experimental goals

- PALEOCCENE – to explore some crystals' potential as a neutron, neutrino, or dark matter detector
- LLNL worked to evaluate crystal damages or color centers after irradiated by various sources
- Existing study hints unstable/permanent damages in some crystals by radiations
- We are looking for the crystal material that is most suitable for a passive neutral particle detection

> - Selectivity of nuclear recoils
> - Visibility of the color centers
> - Stability under environmental hazards



gamma irradiated



neutron irradiated

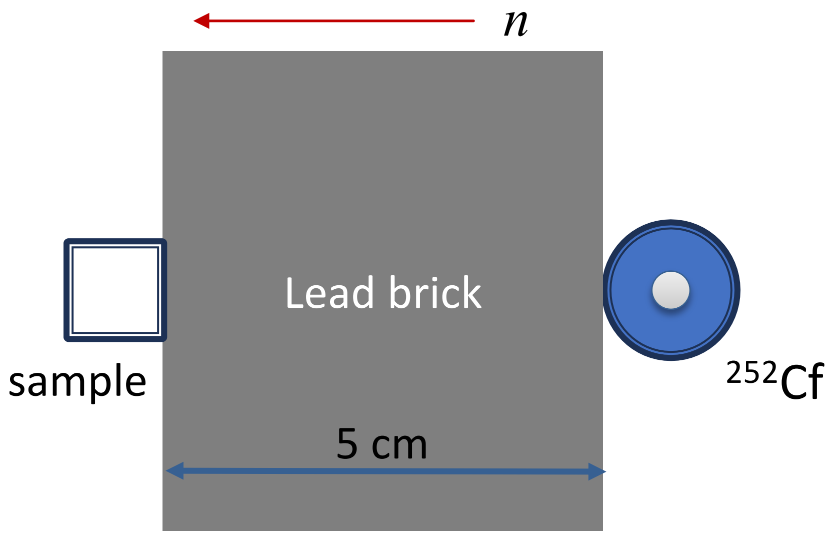
Methodology

- Crystals were exposed to radiation from ^{252}Cf (neutron) and ^{60}Co (gamma)
  - Five samples : Al_2O_3, MgF_2, LiF, CaF_2, BaF_2
- We look for Raman scattering and other spectral features of irradiated samples
  - Fluorescence spectrometer
  - Raman spectrometer
  - Raman microscope (confocal)
- Needs to test the damaged crystals' resistance to environmental hazards
  - Aging
  - Heating
  - UV exposure
- Raman spectra right after the irradiation and after exposure to the hazards were compared

Neutron and gamma irradiation

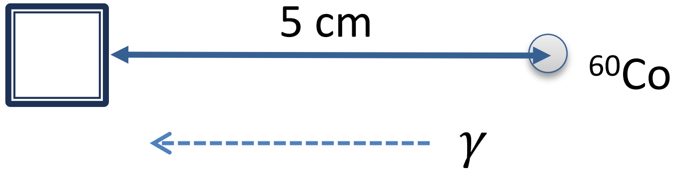
Neutron

- Neutron irradiation done with a ^{252}Cf source (approx. 10^9 neutrons/day/cm^2)
  - Source shielded to block 95% gamma
- Samples exposed to 24 hours of irradiation three times
- Samples tested before irradiation and after each irradiation cycle



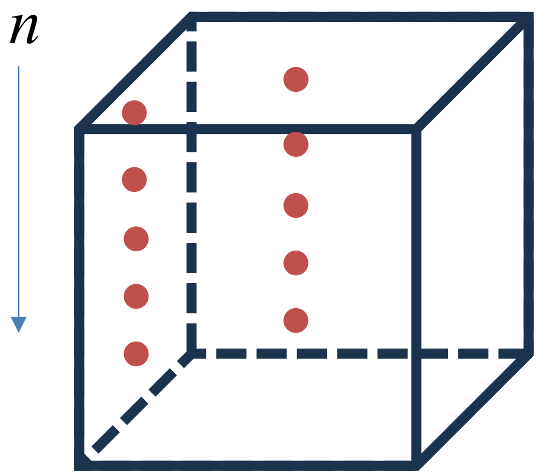
Gamma

- One group was irradiated by ^{60}Co to study the neutron-gamma selectivity
  - (10^{10} gamma/cm^2)

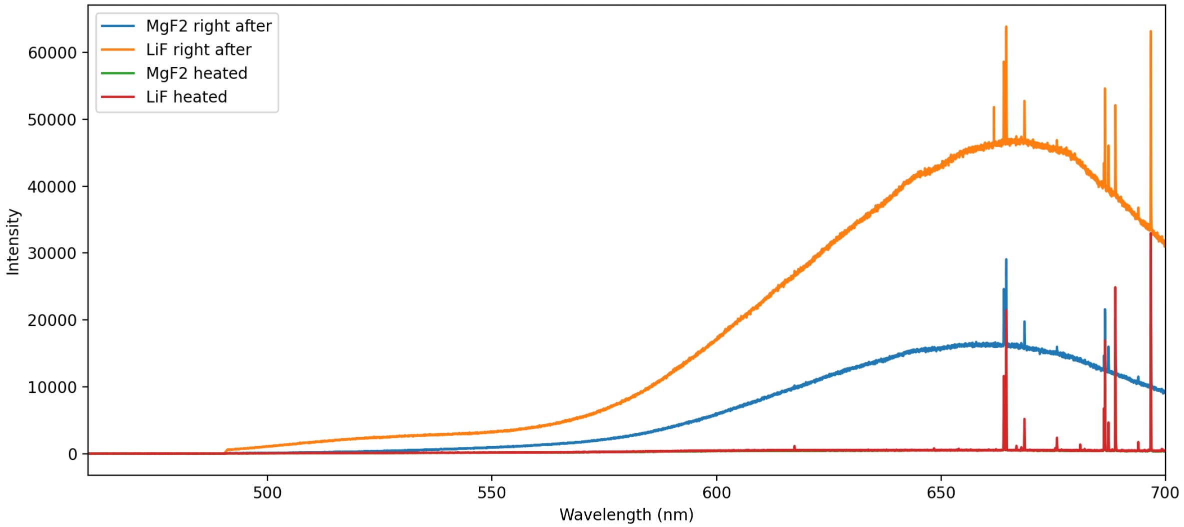


Raman spectroscopy

- Raman spectra were taken at ten different locations in the crystal, from the surface to the center
- Each location has 200 um x 200 um area
- Raman spectra taken in 460 nm to 700 nm range, excited by 457 nm laser



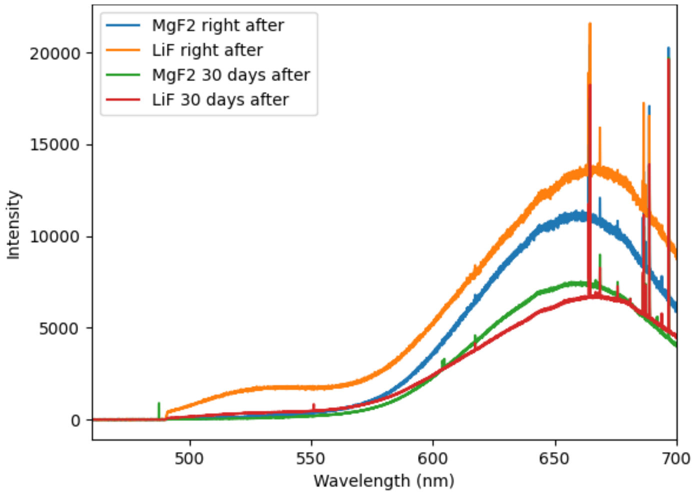
Example of the Raman spectra



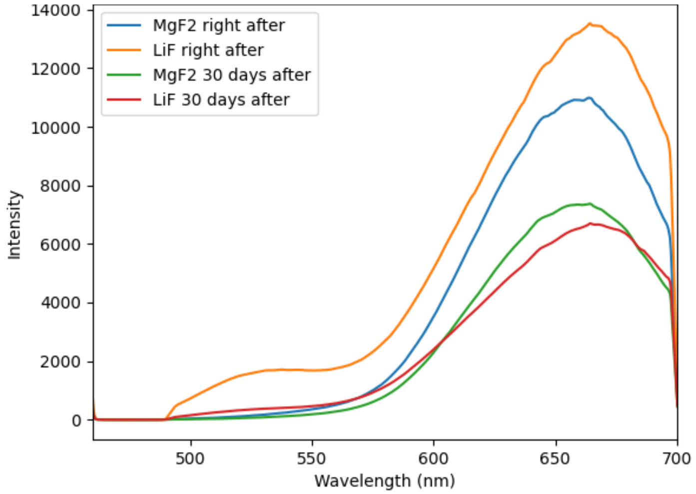
Analyzing the Raman spectra

- The total Raman spectra were fitted with a baseline fitter
- Best fitted baselines are the thermoluminescence (TL) spectra
- Pure Raman peaks are the TL subtracted spectra
  — Raman peaks are summed to represent the change over different conditions

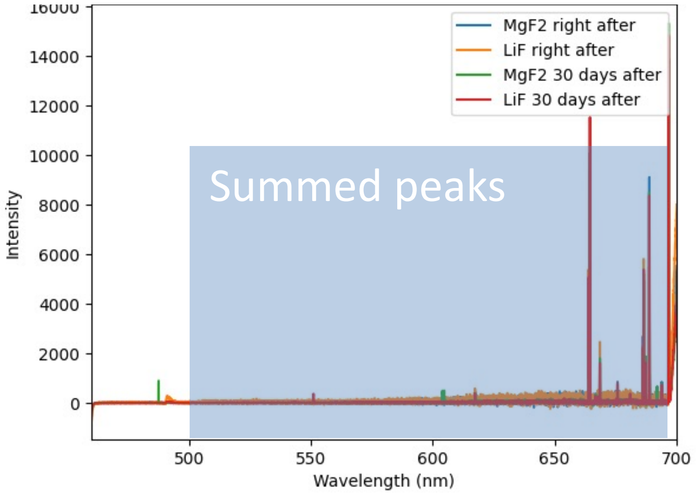
Full spectrum



Baseline spectrum



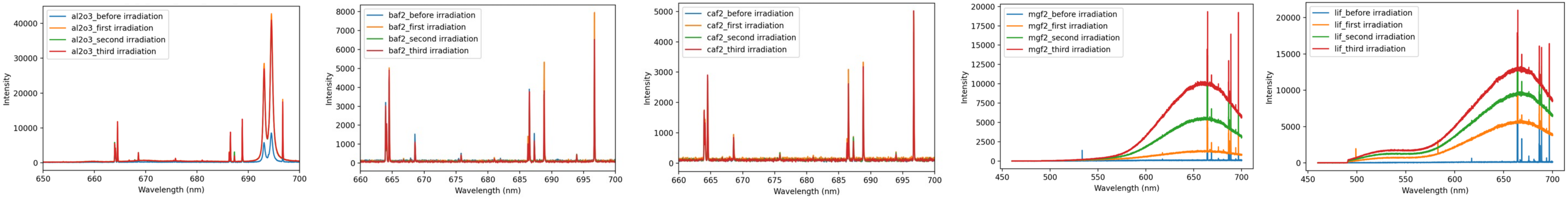
Baseline subtracted



Raman spectra after irradiation

- After three rounds of neutron irradiation, Al_2O_3, MgF_2, and LiF showed most significant Raman spectra changes
- Al_2O_3 changed at two wavelength over the three different periods, but not gradually, nor proportional to the radiation dosage.
- MgF_2 and LiF's spectral change are most significantly TL
- Measuring at single spots in the crystal does **not** guarantee the observation of damage

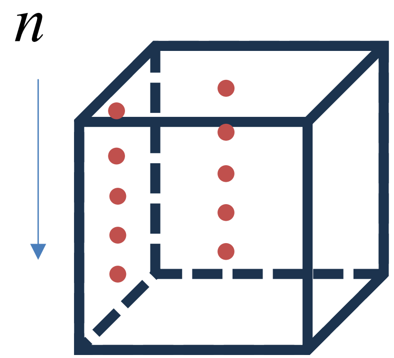
Al2O3 BaF2 CaF2 MgF2 LiF

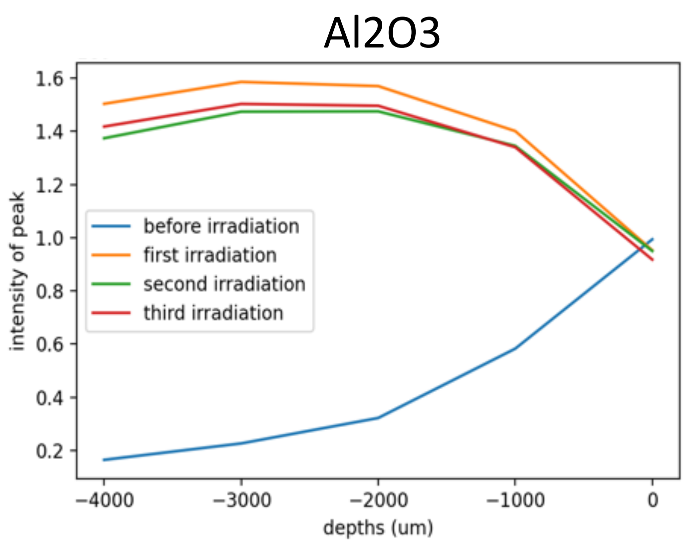


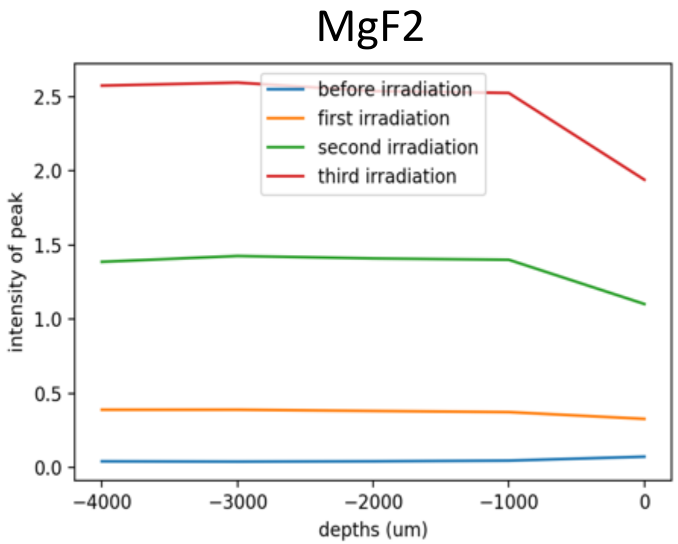
Raman spectra of tested crystals after three 24-hour periods of neutron irradiation.

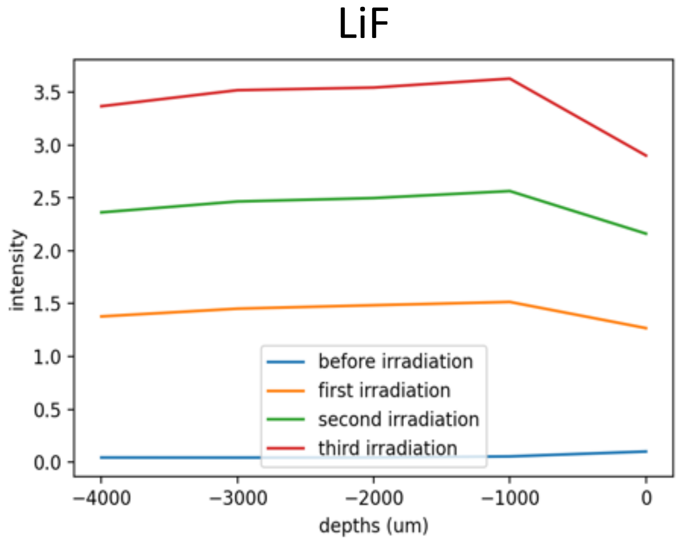
Raman spectra measured at different depths in crystals

- The total Raman intensity is the summed peak intensity in a Raman spectrum
- The total Raman intensity does not show obvious correlations between intensity change and the depths in the neutron travel direction



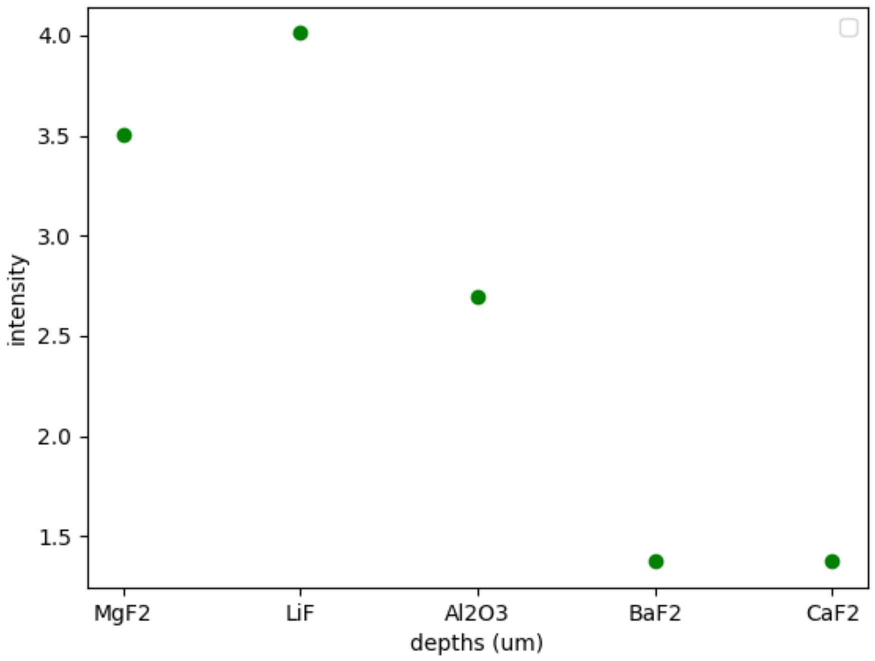






Total light intensity with respect to neutron dosage and depth from locations near the sample bottom to the surface.

Neutron/electron recoil selectivity



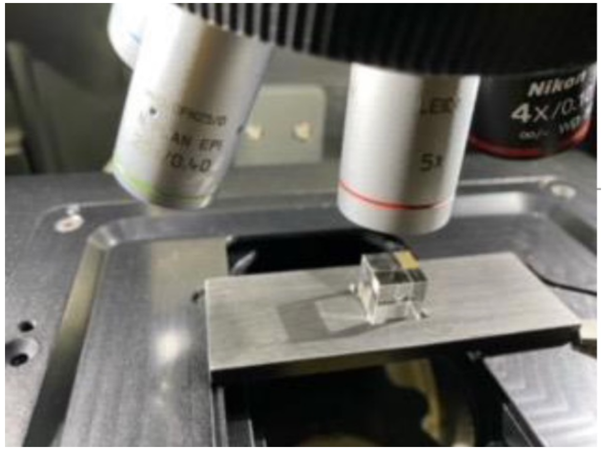
Ratio between neutron and gamma radiated samples' total Raman spectrum intensity.

- Difference between neutron and gamma damages
- 10^9 neutron/cm^2 vs 10^{10} gamma/cm^2 in five samples
- Total Raman spectrum intensity, after subtracting TL baseline, were compared between neutron and gamma irradiated samples
- LiF and MgF_2 showed most significant difference indicating potential good selectivity

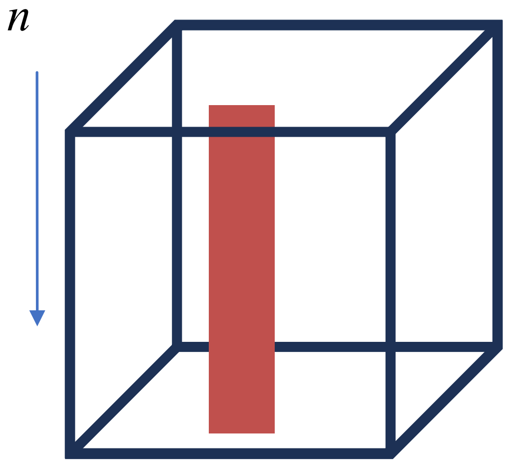
Raman microscopic imaging

- 2D microscopic scan with a confocal Raman microscope
- Measured pixels with 100x100 μm^2 in a 2x10 mm^2 stripe
- Intensities of selected wavelength were measured at each pixel
- The microscopic image can indicate crystal damages at specific wavelengths

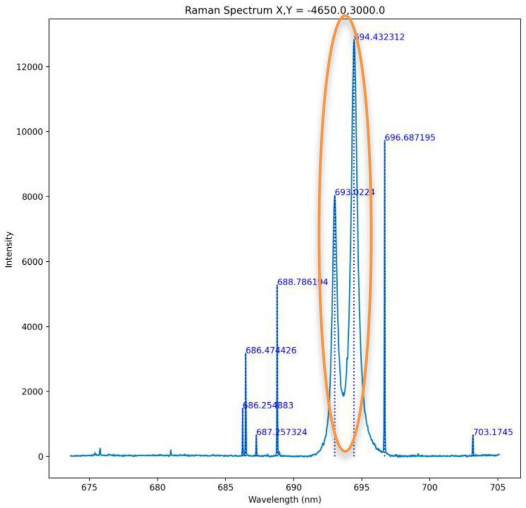
Confocal microscope



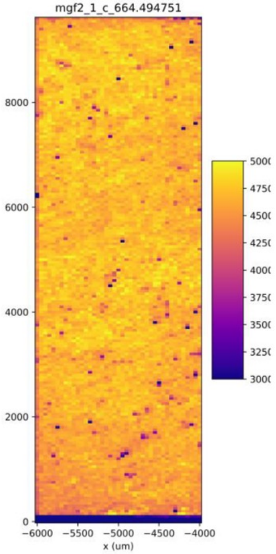
Scan a stripe along the *n* path



Look at a wavelength

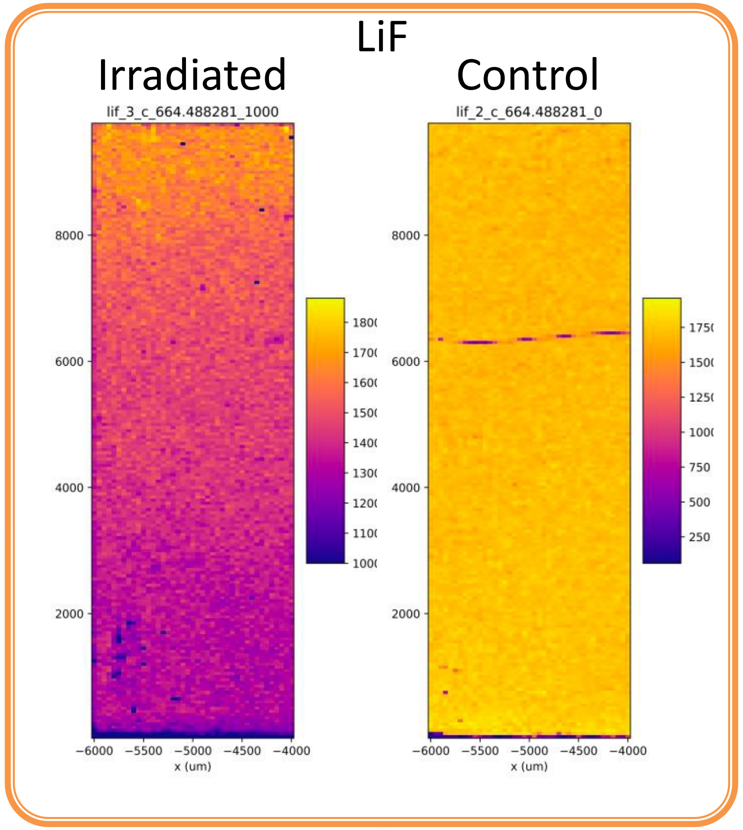


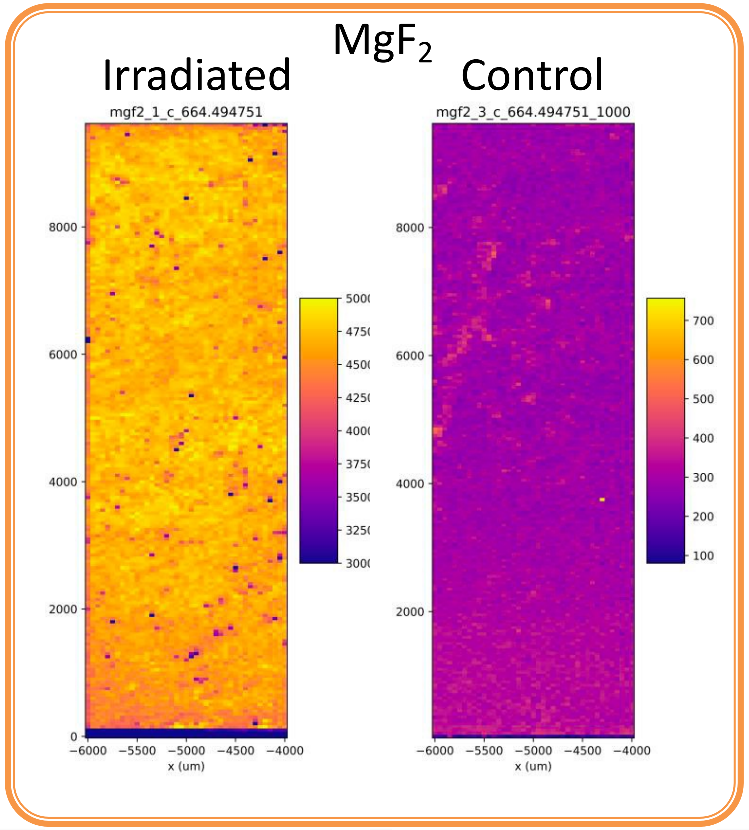
2D image

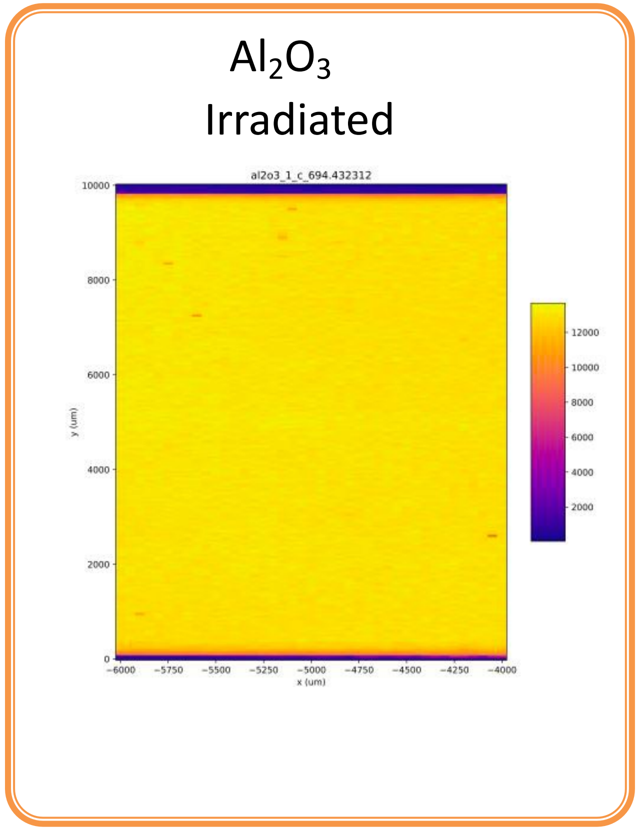


Microscopic images at specific wavelengths

- MgF_2 and LiF both showed dark spots indicating reduced intensity at peaks
- Al_2O_3 does not show significant spots at target Raman peaks

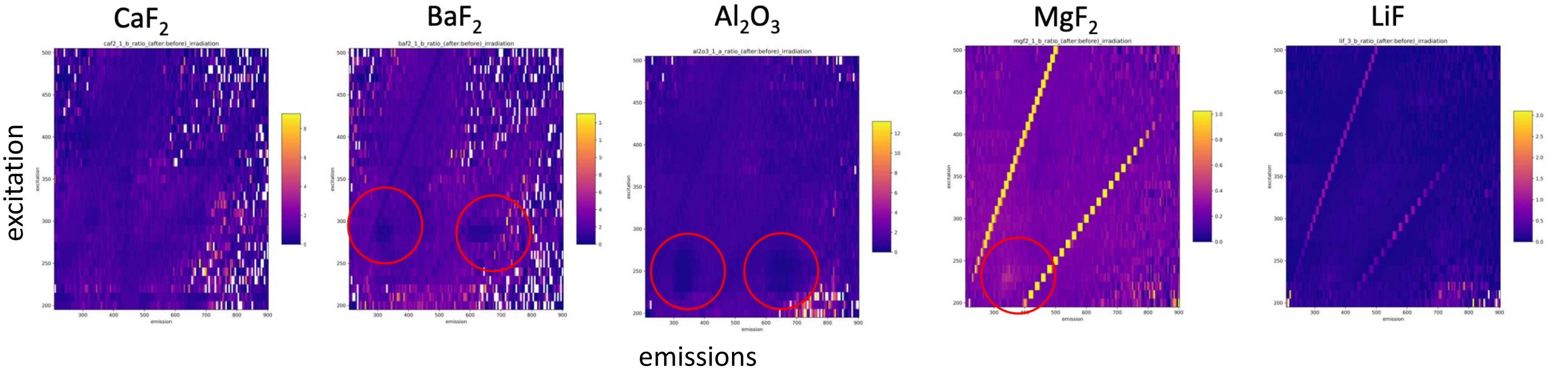






Fluorescence spectra

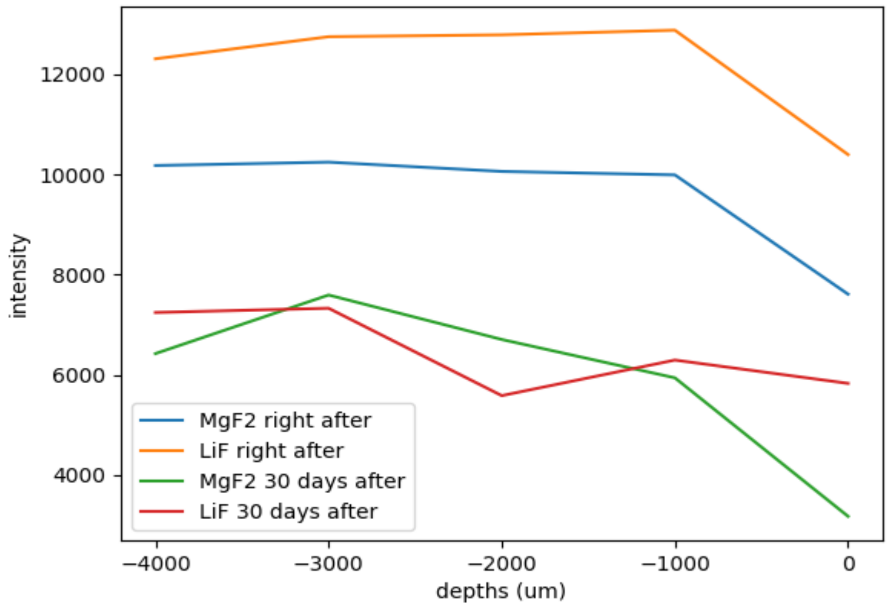
- Irradiated samples were also measured with a fluorescence spectrometer
- The fluorescence measurement is limited with the instrument we used
- Ratio of the fluorescence spectra (irradiated/before)
- Some significant ratio change seen in MgF2, Al2O3 and BaF2



Environmental hazards – crystals after 30-day aging

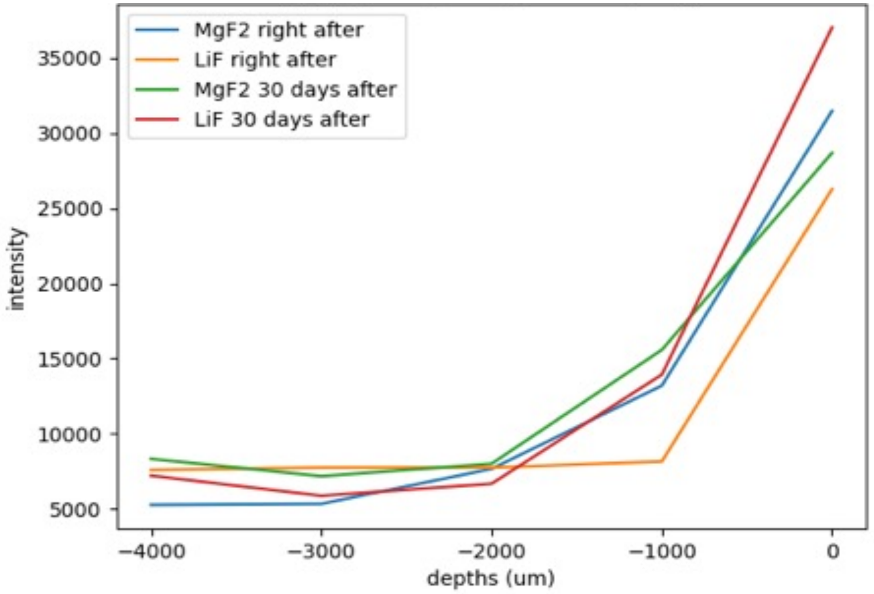
- Three groups of crystal irradiated by ^{252}Cf for 72 hours (6e9 neutrons)
- Aging group – left on shelf in dark, room temperature for 30 days after irradiation before measurement
- LiF and MgF_2 are shown to indicate most significant change

TL light intensities dropped significantly after a month



LiF's slightly recovered Raman intensity

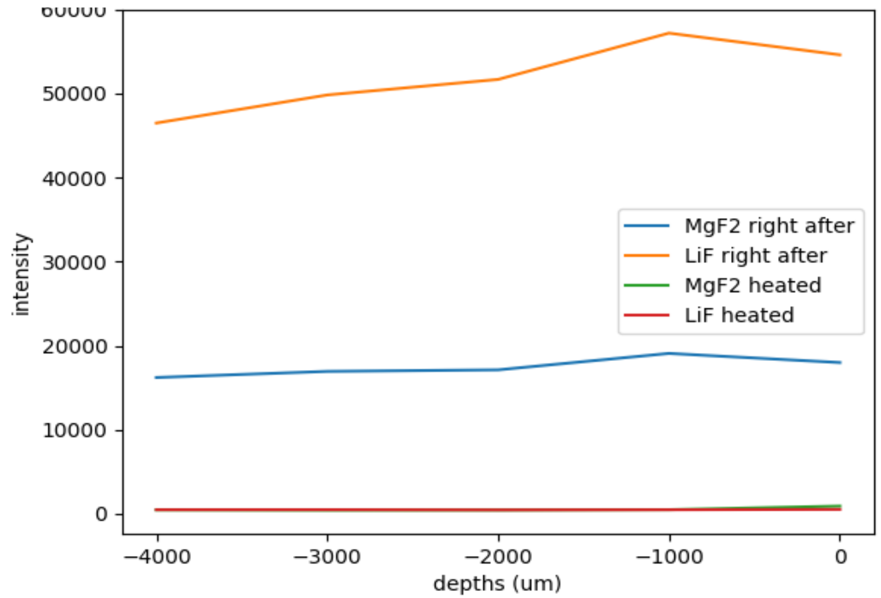
MgF2 remain unchanged



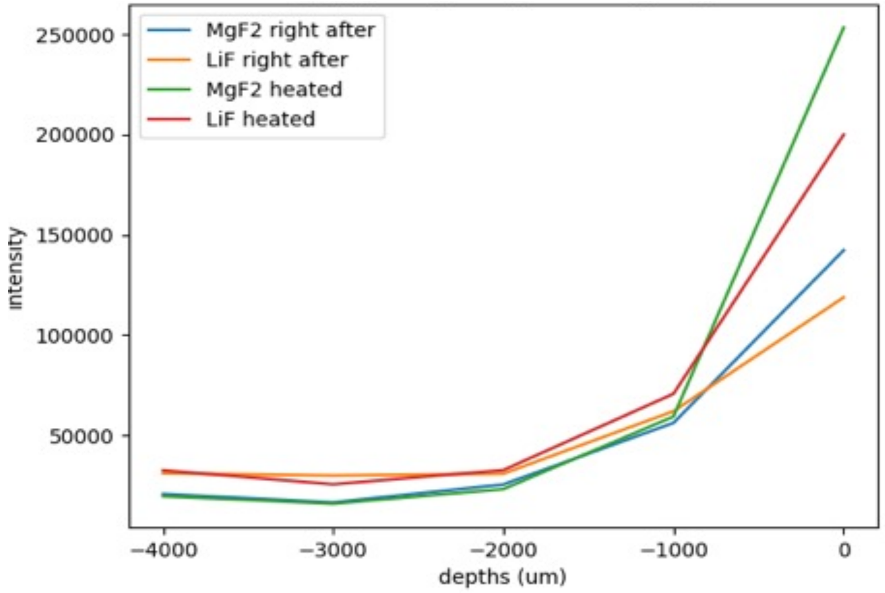
Environmental hazards - crystals after 500 °C heating

- Heat group – baked in a furnace with 500 °C for 1 hour (heating and cooling rate 5 °C/s)
- Melting points are above 1000 °C

TL in samples were eliminated after heating

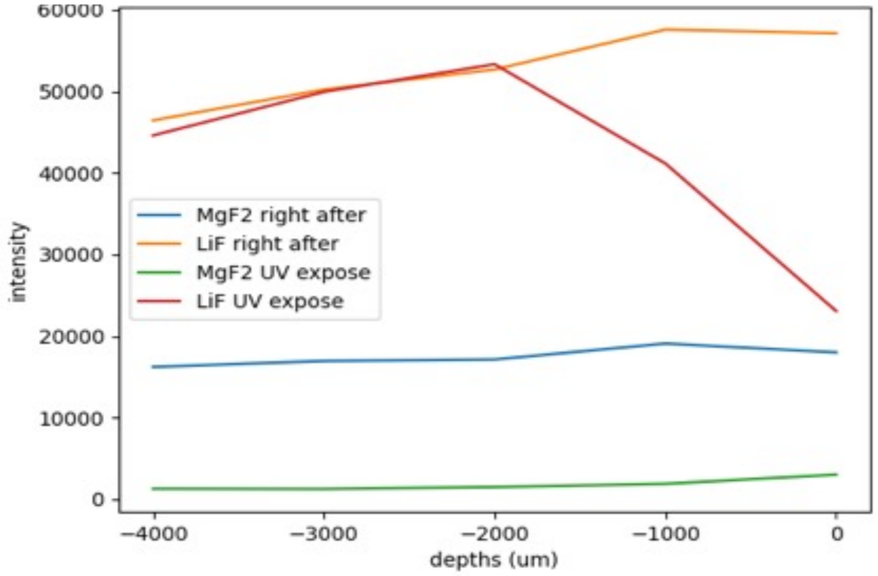


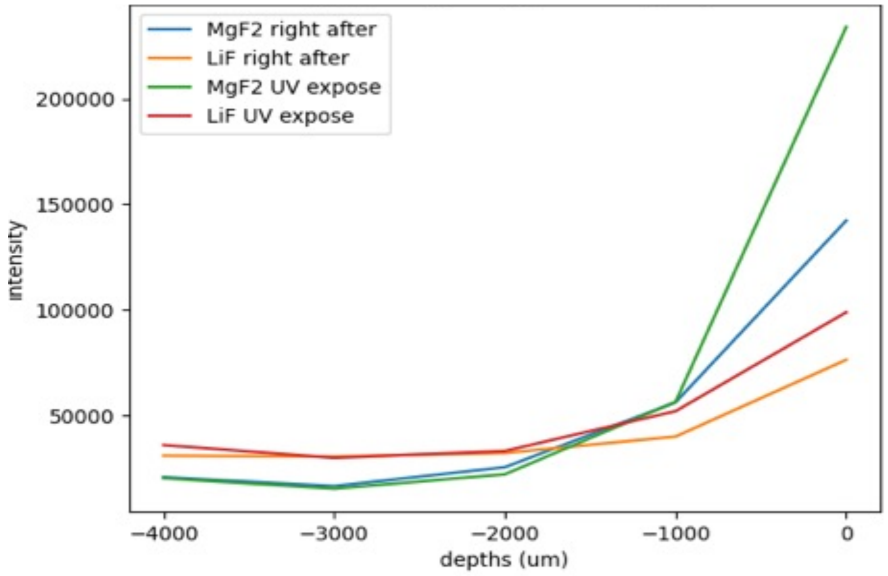
Total Raman peak intensities recovered more significantly near the surface of crystals



Environmental hazards - crystals after 24hr solar UV exposure

- UV group – exposed to sunlight (average UV index 8) for 24 hours at 28 °C temperature
  - Equivalent UV intensity 0.2 mW/m^2
- TL light intensity reduced more near the surface of LiF after UV, greatly reduced in MgF_2
- Raman peak intensities recovered more significantly near the surface of crystals, for both materials



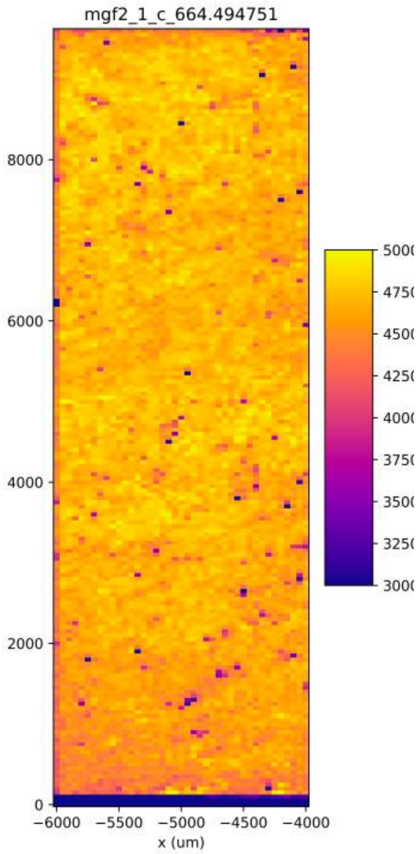


Discussions

- The study currently is limited by the instruments available.
  - The Raman microscope taking one pixel per scan is the bottleneck
  - Measurement on a 1 mm x 5 mm area takes four hours
- Need to zoom in to investigate the dark spots
- A SPIM system would significantly improve the measurement quality and efficiency
- Characterizing the energy response of the crystals is the next major milestone
  - LLNL is capable to irradiate samples with various energies

Summary

- Crystals were irradiated by neutron and gamma sources
- We characterized the crystals using Fluorescence and Raman spectrometry
- LiF and MgF_2 had dark spots in 2D Raman microscopic images after irradiation
- We did aging, heating, and UV exposure to three groups of neutron irradiated crystals
- TL and pure Raman peaks were looked separately in the spectra comparisons
- The environmental effects are more significant near the surface of the crystal, which are more easily impacted by heating and UV exposure



Backup – general knowledge

- Electron and nuclear recoils cause crystal damages
- Some of the damages can be measured as color centers (CC)
- Nuclear recoil caused damages can used to detect neutrons, neutrinos and dark matters
  - Neutron recoils
  - Coherent Elastic Neutrino-nucleus Scattering (CEvNS)
  - WIMP scattering with nuclei
- Some crystals, such as Al2O3, shown to be capable to intrinsically discriminate electron and nuclear recoils in their optical spectra
- We conducted tests to show how potential crystal targets response to neutron radiations

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