Ultrafast Laser Irradiation as a Surrogate for Swift Heavy Ion Irradiation of Actinide Materials

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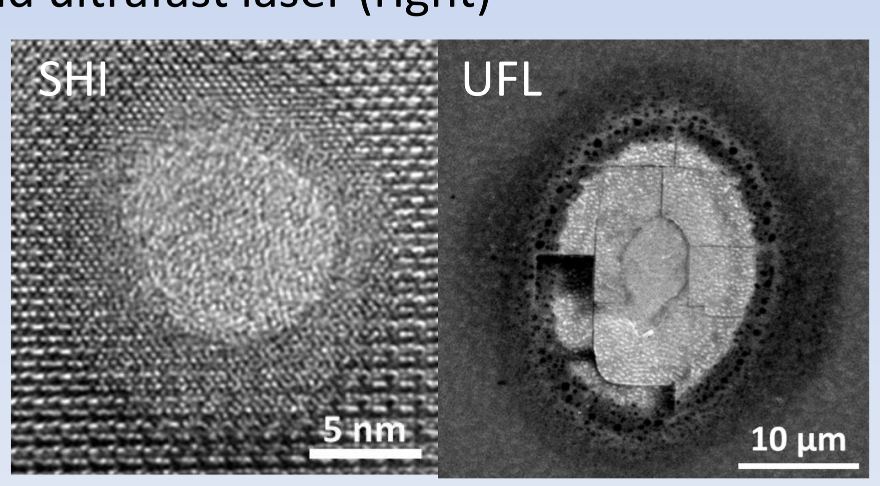
Motivation

- Need to understand response of actinide compounds and actinide-bearing materials to SHI irradiation
- SHI irradiations require access to limited facilities
- Ultrafast laser irradiation can be done at the bench-scale in most laboratories
- Both techniques induce intense electronic excitation
- We are investigating the use of ultrafast lasers instead of SHIs for the study of actinide materials in extreme environments

Energy Deposition

- Swift Heavy Ion:
  - Deposits energy primarily through inelastic collisions with electrons
  - Caused by ion's point charge electric field
  - Creates tracks 3-10 nm wide, microns in length
- Ultrafast Laser
  - Deposits energy through multiphoton + tunneling ionization → inverse Bremsstrahlung → avalanche ionization
  - Caused by laser's oscillating electric field
  - Creates damage spots 10's of microns wide, 10's to 100's of nm deep
- Representative morphologies following irradiation by SHI (left) and ultrafast laser (right)



Damage Process

- Promotion of electron by incident radiation on fs timescales
- Alteration of interatomic potentials on sub-ps timescales
- Electron-phonon coupling from 1-10 ps
- Shock wave propagation from 20-100 ps

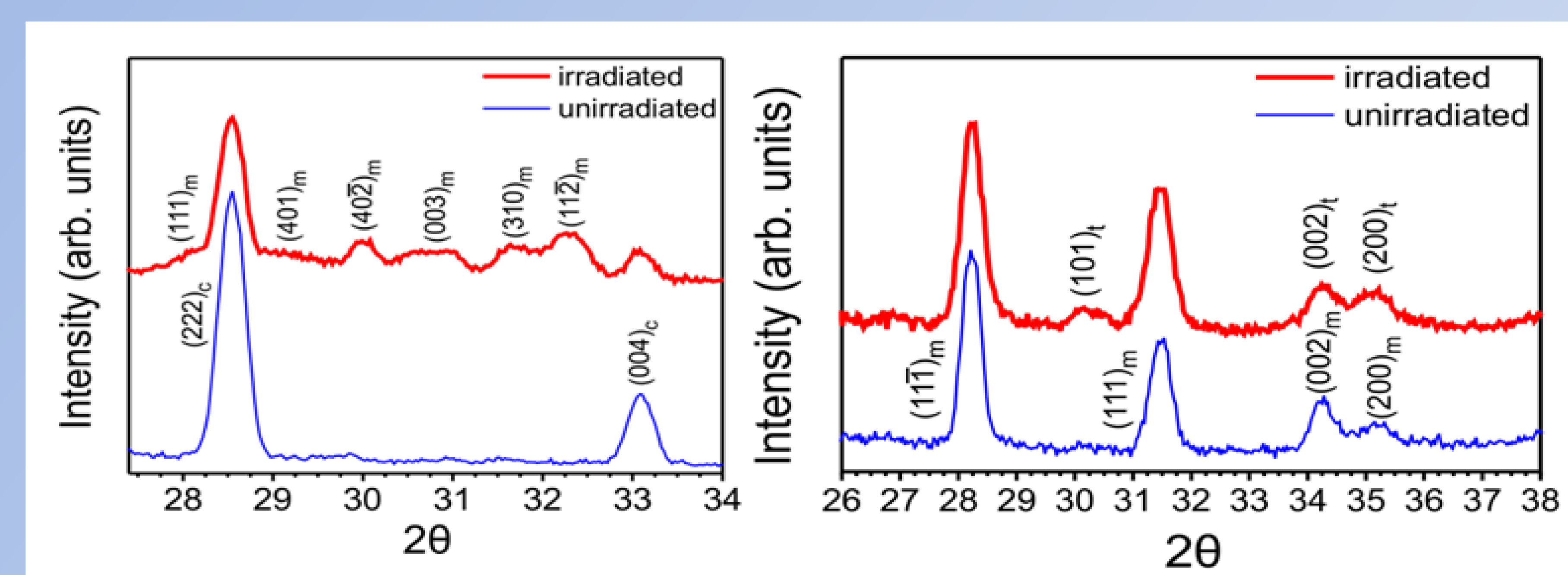
Phase Characterization

Phase transformations match results from SHI irradiations

cubic-to-monoclinic transformation in Gd_2O_3

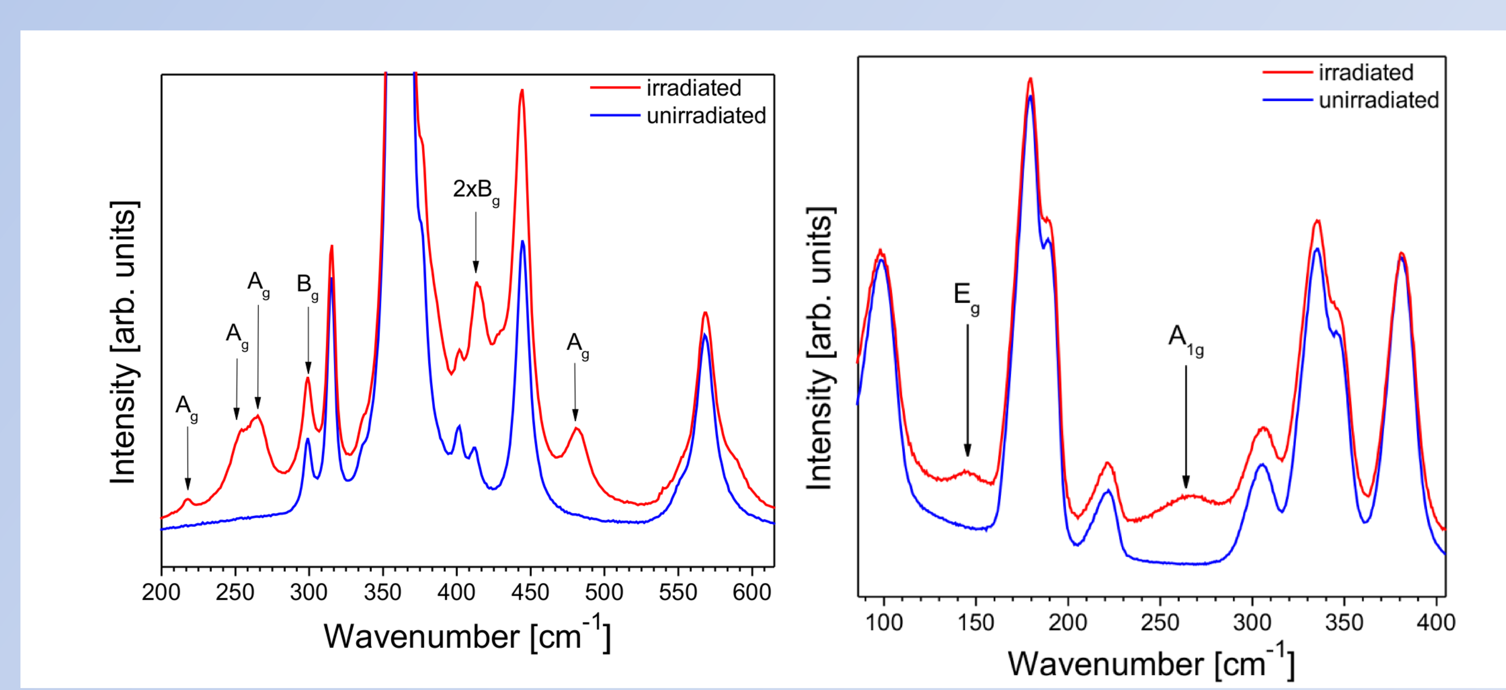
monoclinic-to-tetragonal transformation in ZrO_2

Grazing Incidence X-Ray Diffraction



- Irradiation at 40 J/cm^2
- GIXRD shows that transformation occurs and materials retain long-range order
- Presence of original phase in irradiated spectra indicates incomplete transformation or x-ray penetration depth greater than laser-induced transformation depth
- Raman spectroscopy required to further differentiate between identified phases and other high temperature and high pressure polymorphs

Raman Spectroscopy



- Irradiation at 40 J/cm^2, 10 shots for Gd_2O_3 and 0.5 J/cm^2, 1,000 shots for ZrO_2
- Raman spectroscopy confirms phase transformations
- No phase transformation detected in ZrO_2 at high fluences which indicates a loss of short range order due to defect accumulation

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Damage Mechanisms

Does irradiation cause melting?

High Fluence Regime

- Irradiation Fluence > Melting Threshold Fluence
- Thermal Effects
  - Two-temperature model (Thermal Spike)
  - Electron-phonon coupling causes localized melting
- Shock Wave Generation
  - Expansion of molten material causes shock wave
  - Coulomb explosion possible
- Enough energy to rearrange structures

Low Fluence Regime

- Irradiation Fluence < Melting Threshold Fluence
- Bond weakening
  - Allows for atoms to move from lattice sites
  - Solid state damage mechanism
- Sub-melt threshold means structure is unlikely to rearrange from thermal or shock wave effects

Conclusions

- Phase transformations caused by ultrafast laser irradiation matches results from SHI irradiations
- Positive results have been shown in multiple materials
- Similarity in damage mechanisms provides novel ways to study SHI-material interaction

Future Work

- Extend work to actinide compounds and actinide-bearing materials
- Use ultrafast laser to rapidly screen novel materials before more complicated SHI irradiations
- Use ultrafast laser to further study and isolate mechanisms underlying SHI damage
- Utilize facilities at SLAC to probe excited materials at atomic scale with sub-picosecond time resolution