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Low Energy Ion Irradiation and Its Applicability to Mimic Materials Irradiation Damage from High Energy Protons

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HPT R&D Roadmap Workshop

May 31 - June 1, 2017, Fermilab

Driving Force for Microstructural Changes Induced by MeV Ion Irradiation in Solids

1. Nuclear energy deposition:
   Elastic collision, damage cascades
2. Electronic energy deposition:
   Electron excitation, ionization
3. Electron-phonon coupling:
   Heat production, temperature increase

Emulation of Microstructural Features Using MeV Ion Irradiation and Thermal Annealing

Benefits:

- ☺ Accurate dose for emulation of material age
- ☺ Accurate temperature for emulation of the location inside the material with a temperature gradient
- ☺ Minimum or no radiological activation for immediate release and characterization of irradiated materials.
- ☺ Implantation of impurity species into a pre-existing structure without thermal constraints.
- ☺ Fast emulation of structural features within hours to days
- ☺ Low cost

Limitations:

- ☹ High dose rate
- ☹ Possible temperature shift

MeV Ion Irradiation to Emulate High Energy Proton Irradiated High Power Target Materials

- Irradiation damage in HPT materials starts from production of point defects, followed by their accumulation and interactions, leading to formation of defect clusters up to full amorphization.
- Point defects are produced mainly by irradiation of spallation neutrons and ions, especially at low energies, which may be emulated by low energy ion irradiation.
- The effects of temperature and its possible gradient in HPT materials may be emulated through post-irradiation thermal annealing at high temperatures, which may lead to formation of fractures and cracks.
- Gas bubbles and solid state precipitates in HPT materials may be emulated by implanting the species.
- Each contributor may be emulated separately or in a combined way to some extent.

Proposed Procedure to Emulate High Energy Proton Irradiated High Power Target Materials

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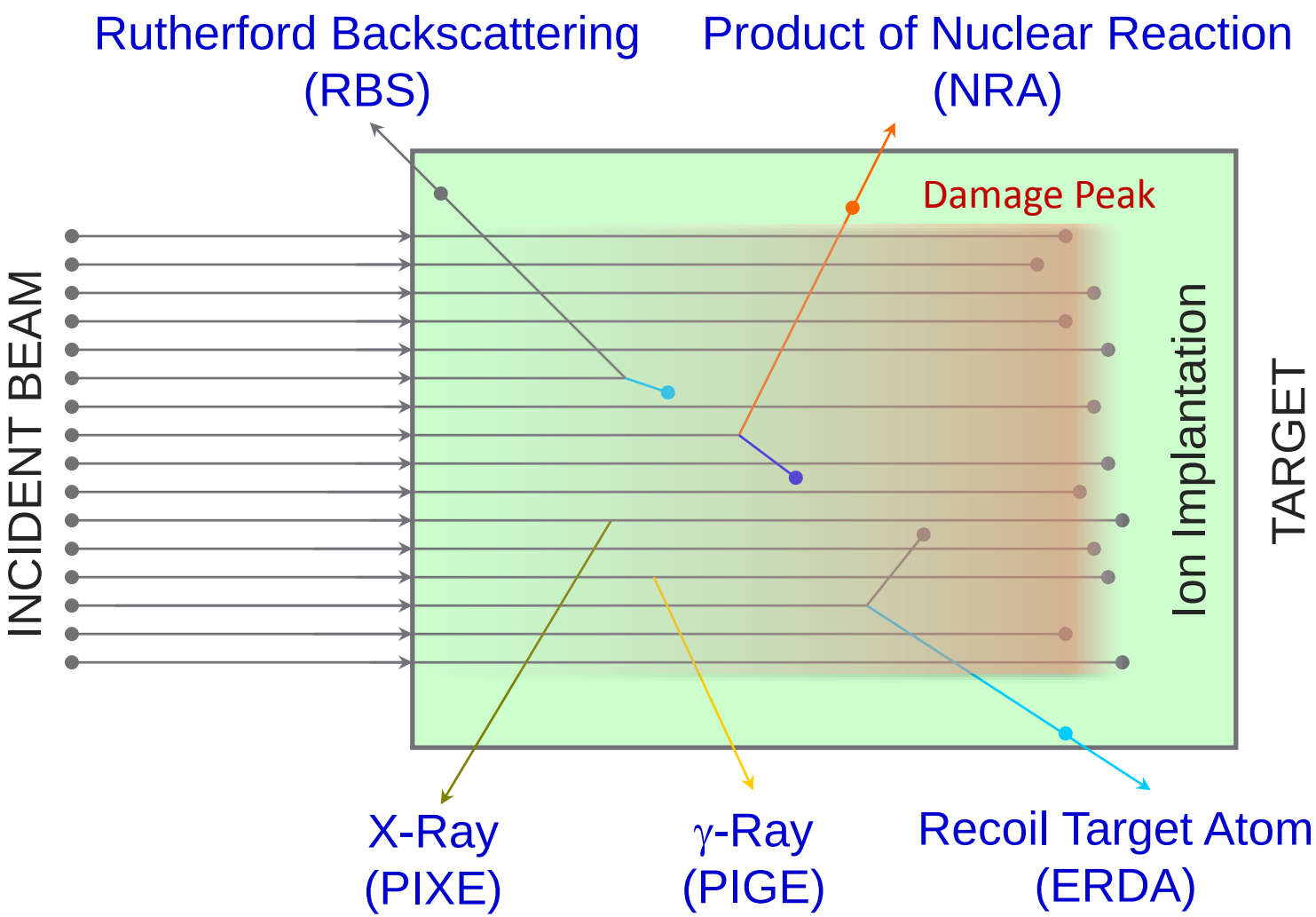
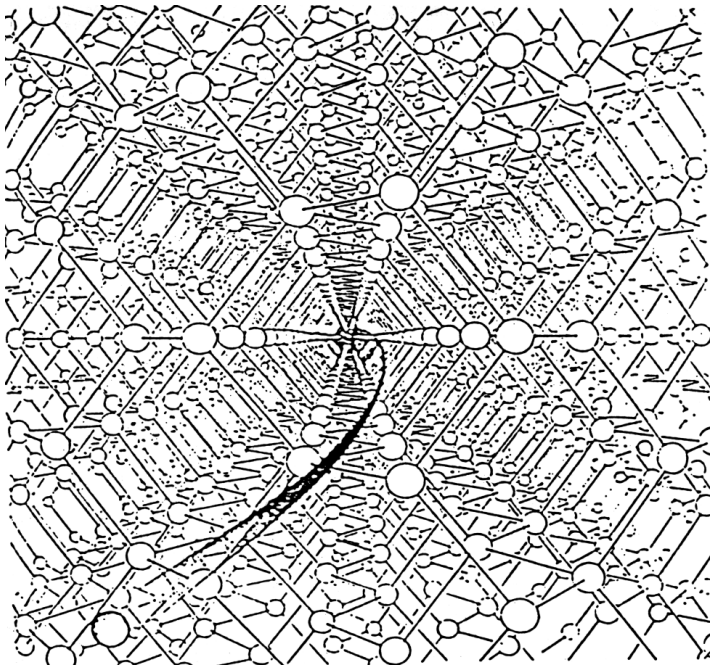
- To simplify data interpretation, start with highly oriented pyrolytic graphite (HOPG), pure light metals or model alloys without grain boundaries, pores or high-level impurities, followed by polycrystalline materials with increasing levels of material complexities.
- Perform in-situ damage accumulation study of HOPG irradiated, for example, with H^+ ions and self-ions (C^+) as a function of dose and temperature.
- Perform in-situ and ex-situ thermal annealing study of defect recovery and clustering.
- Perform in-situ HIM irradiation study of microstructural evolution in polycrystalline graphite.
- Perform microscopy study of HOPG and polycrystalline graphite implanted with H, He and non-gaseous spallation/transmutation species (e.g., Li) and annealed at high temperatures to emulate microstructures for study of various features, including polycrystallization, amorphization, shrinking/swelling, creep, Mrozowski cracks, gas bubbles, and precipitates.
- Measure physical properties, including thermal conductivity, electrical conductivity, and mechanical strength.
- Compare the emulated microstructures and properties with those of high energy proton irradiated graphite and develop a fundamental understanding of the structure-property relationships, which may help assess and predict material performance.

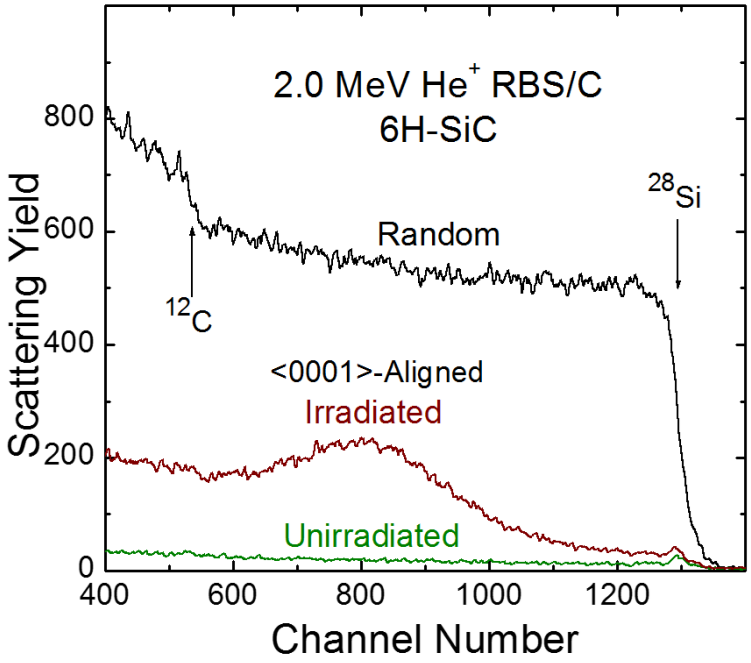
Fundamental Processes of Ion-Solid Interactions in the MeV Energy Range



Ion Channeling and RBS/C

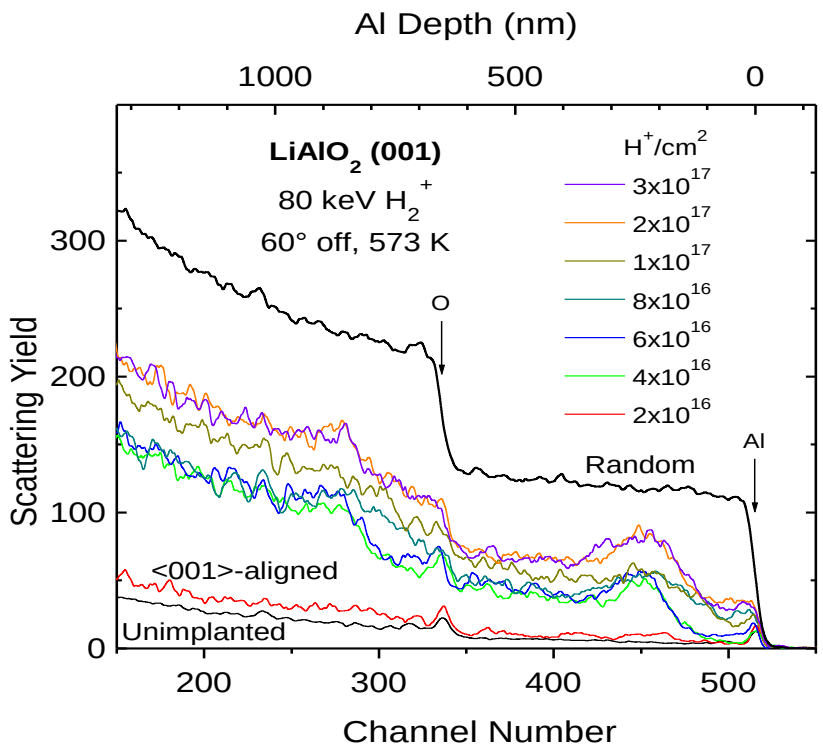


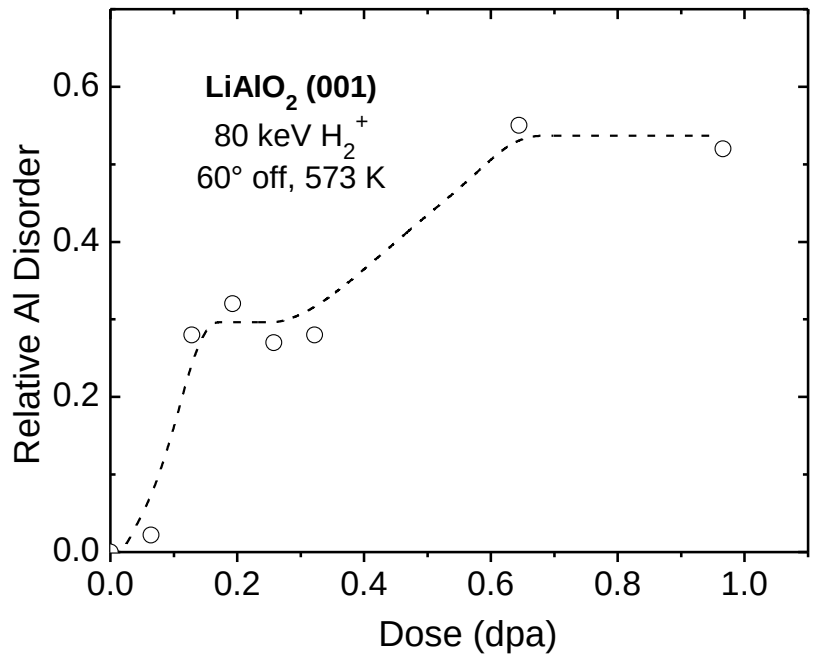
From L. C. Feldman, et al.,
"Materials Analysis by Ion Channeling"



RBS/C and random spectra for 6H-SiC

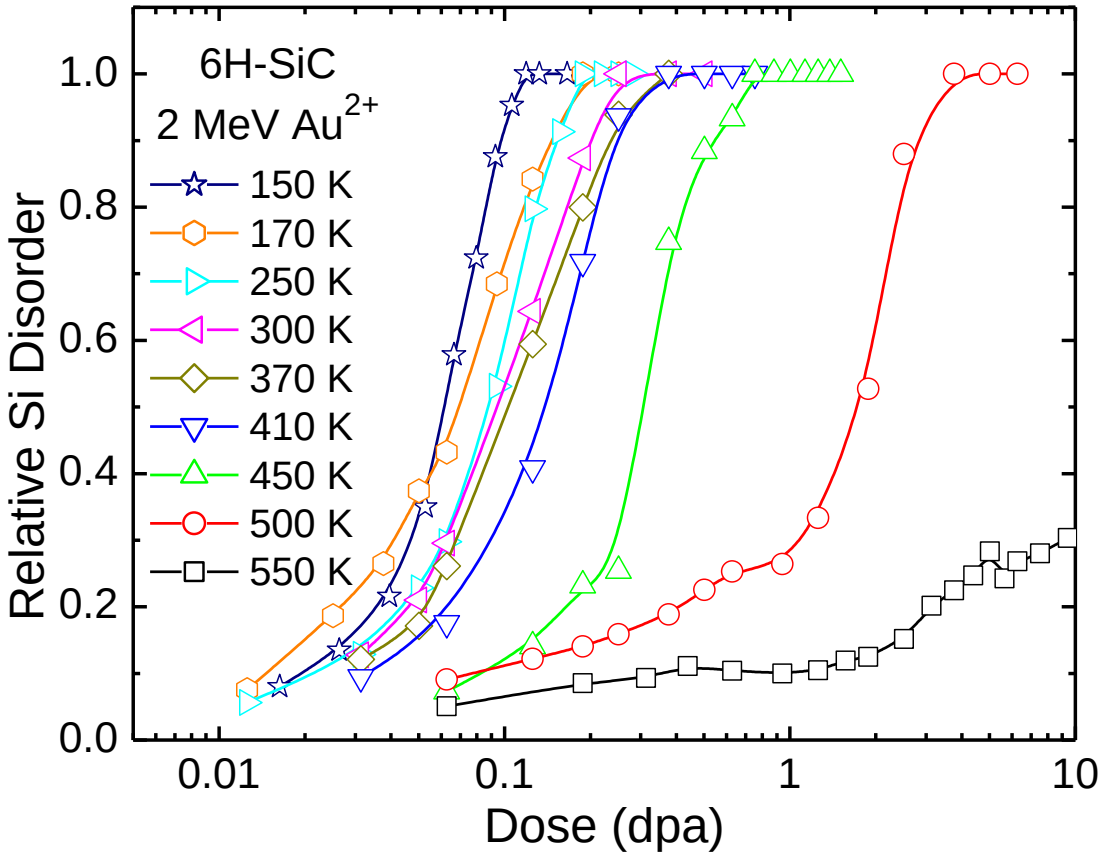
Disorder Accumulation in γ-$LiAlO_2$ at 573 K





- Disorder on the Al sublattice saturates at levels of 0.3 and 0.5.
- No full amorphization occurs at the highest applied dose of 1 dpa.

Effect of Irradiation Temperature on Disordering Rate



Disordering rate decreases with increasing irradiation temperature due to simultaneous recovery

Thermal Recovery of Defects on Both Si and C Sublattices in Irradiated SiC

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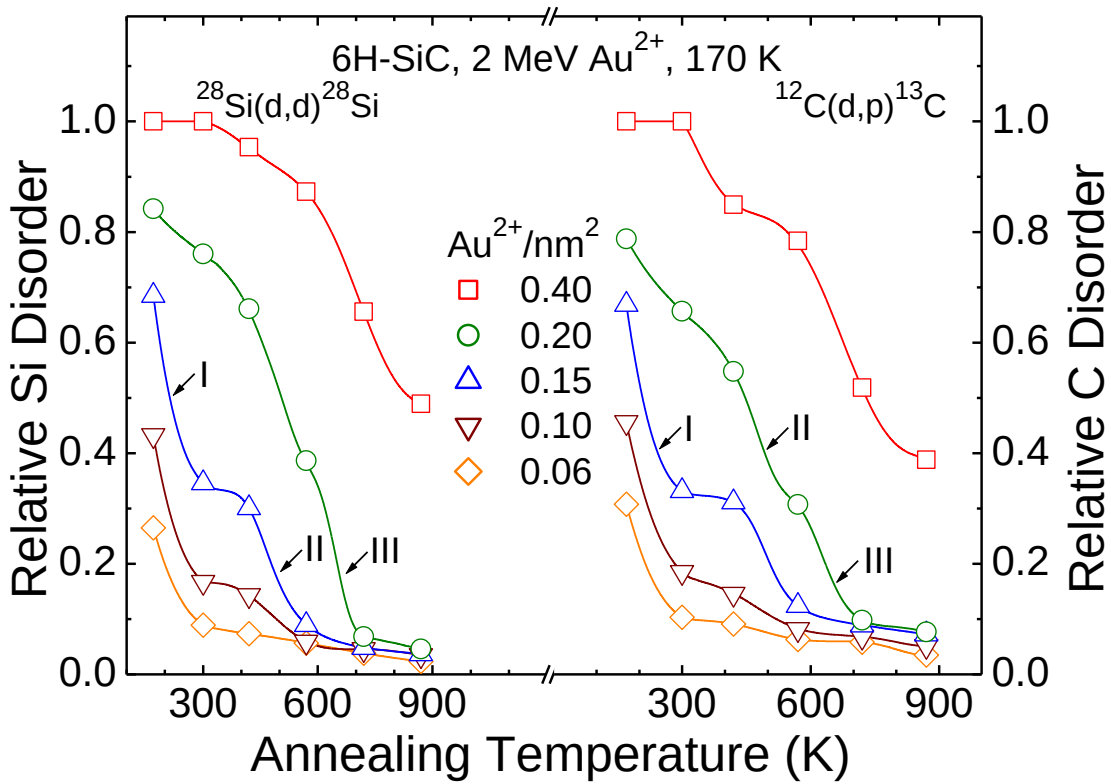
20-min Isochronal Anneals



Similar recovery stages (I, II, III) on both Si and C sublattices

Li and H Out-diffusion in H^+ Irradiated γ-$LiAlO_2$

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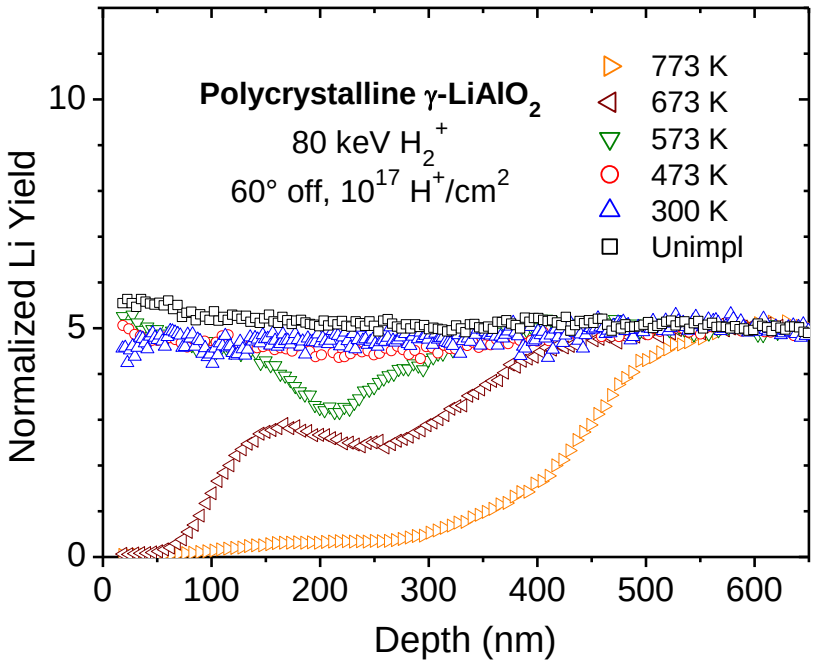
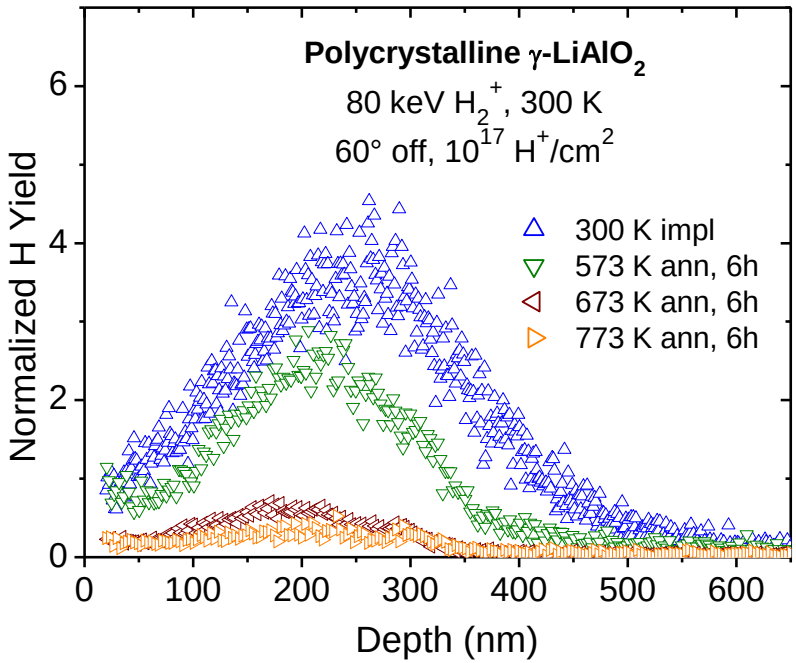
During H^+ Implantation



During Thermal Annealing



- Material decomposition, Li diffusion and loss during irradiation
- H diffusion and release during thermal annealing

Amorphization and Precipitate Formation

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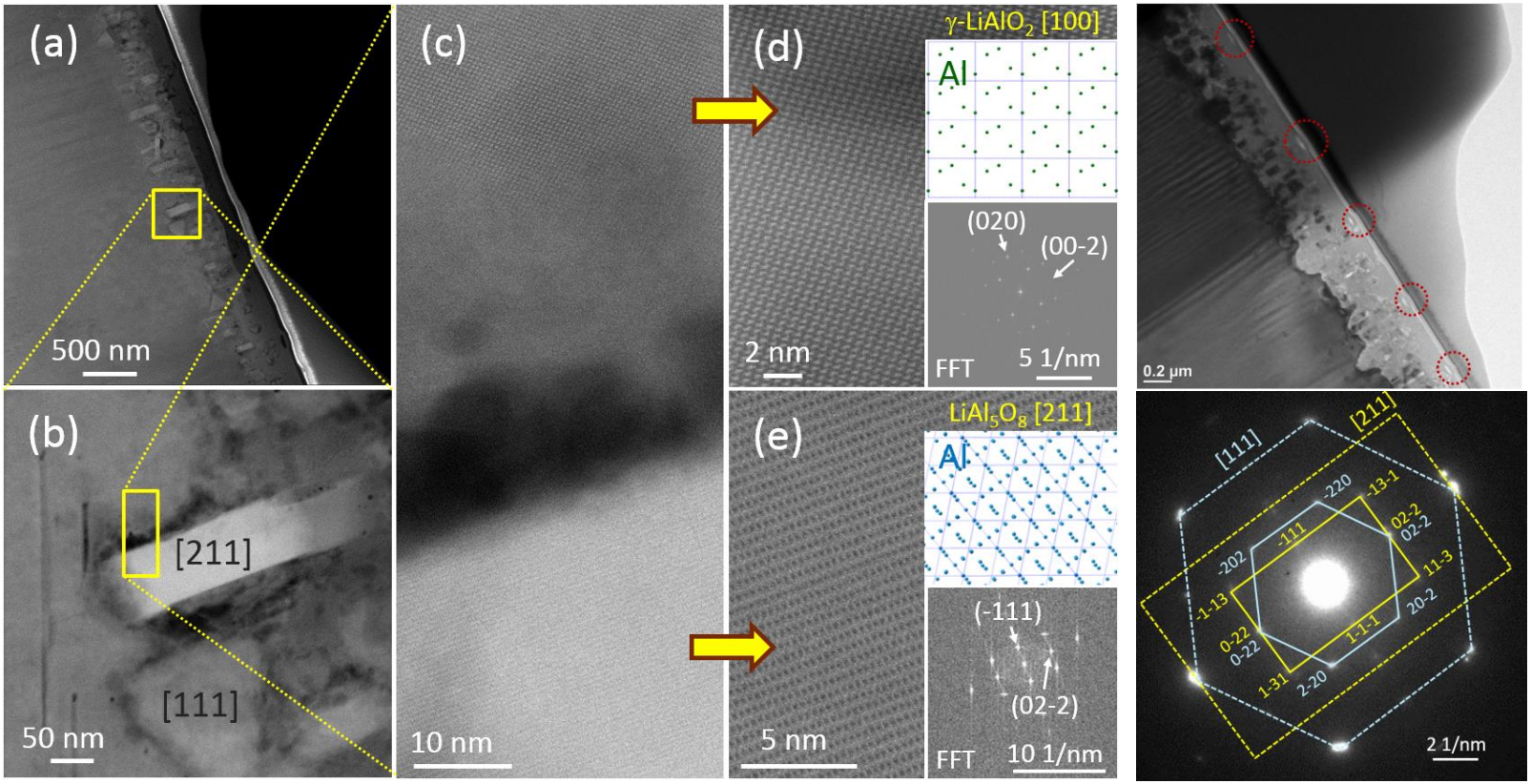
γ-$LiAlO_2$ implanted to 10^{17} H^+/cm^2 at 773 K



- The precipitate in rectangular shape is identified as cubic $LiAl_5O_8$ with zone axis [211] that is parallel to γ-$LiAlO_2$ [100].
- Precipitates also show in triangular shape, which has a zone axis [111].
- Amorphization and gas bubbles near the surface are observed.

STEM-EELS Mapping of Precipitates in 3C-SiC

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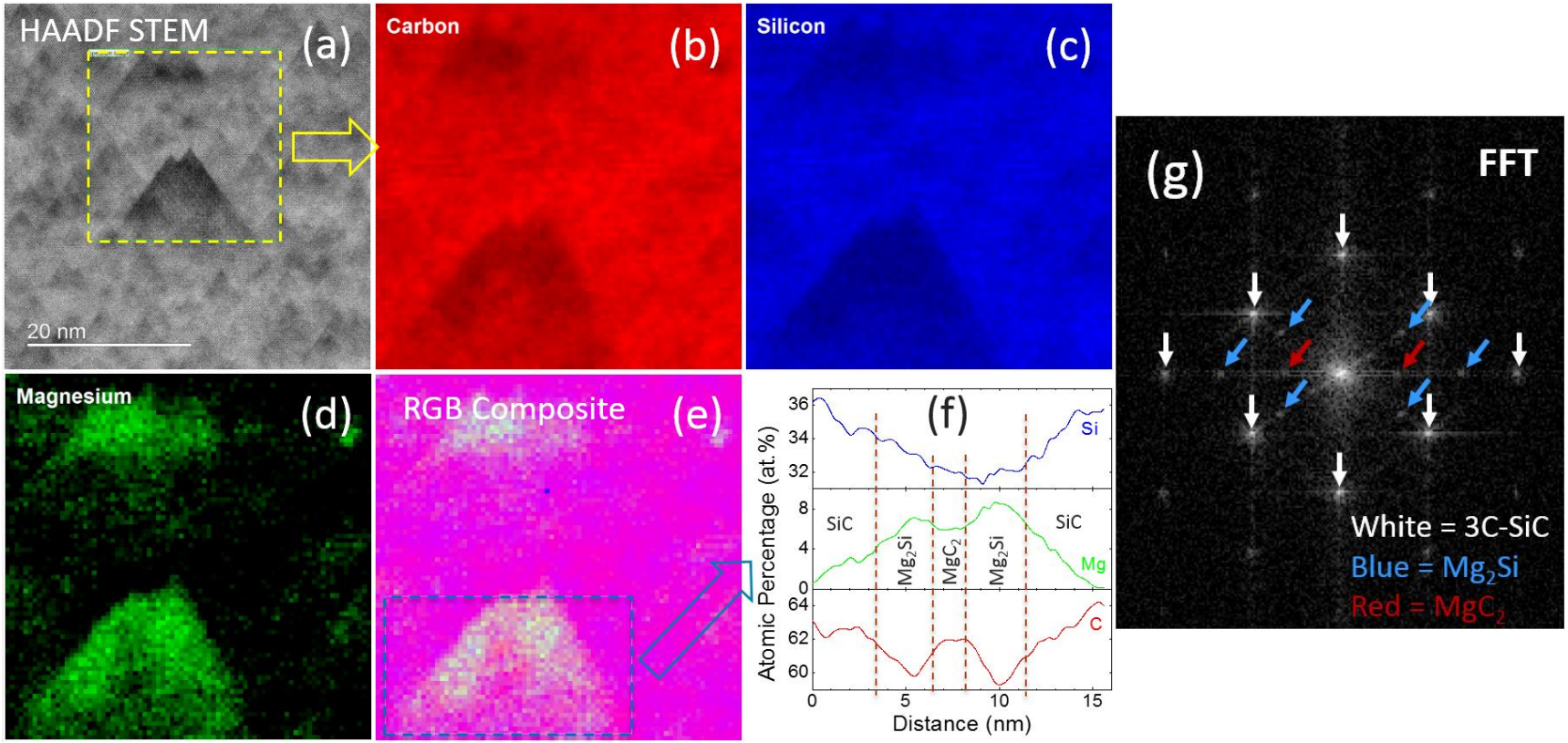
3C-SiC implanted to 9.6×10^{16} $^{25}Mg^{+}/cm^{2}$ at 673 K and annealed at 1573 K for 12 h



Formation of cubic Mg_2Si and tetragonal MgC_2 tetrahedra in Mg^{+} implanted 3C-SiC.

Helium Ion Microscope (HIM) at PNNL/EMSL

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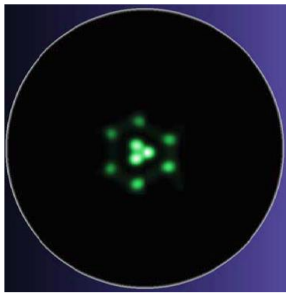
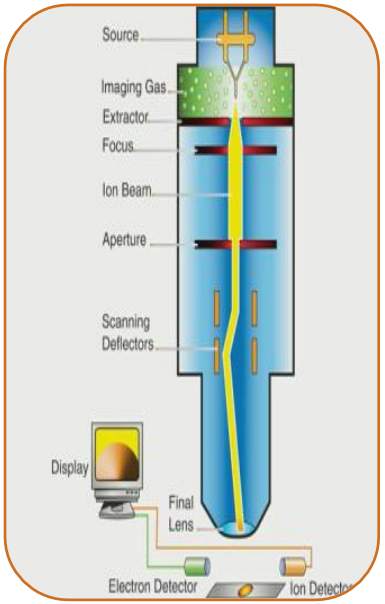
Source Tip Trimer



The Column



As an advanced instrument, HIM was developed and commercialized in 2007, providing cutting-edge imaging and chemical analysis with a sub-nanometer probe. <u>One of the unique capabilities is the in-situ study of microstructural evolution in bulk material at a microscopic site of choice under He^+ ion irradiation.</u>

Specifications and Capabilities

- Small beam size: < 0.1 nm
- High resolution: ≤ 0.35 nm
- Magnification: 100 – 1,000,000
- Field of view: 1 mm – 100 nm
- Depth of field: 5-7 times SEM
- RBS spatial resolution: ~10 nm
- Variable voltage: 5 – 30 kV
- Beam current: 1 fA – 25 pA
- No conductive coatings needed
- High surface sensitivity
- High image contrast
- Low Z imaging
- Backscattered ion imaging

Examples of Applications

- Nanostructures in nuclear materials
- Precipitates, gas bubbles, grain boundaries, cracks, interfaces, etc.
- Irradiation modification of material structures using sub-nanometer He^+ ion probe

He Bubble Formation in γ-$LiAlO_2$

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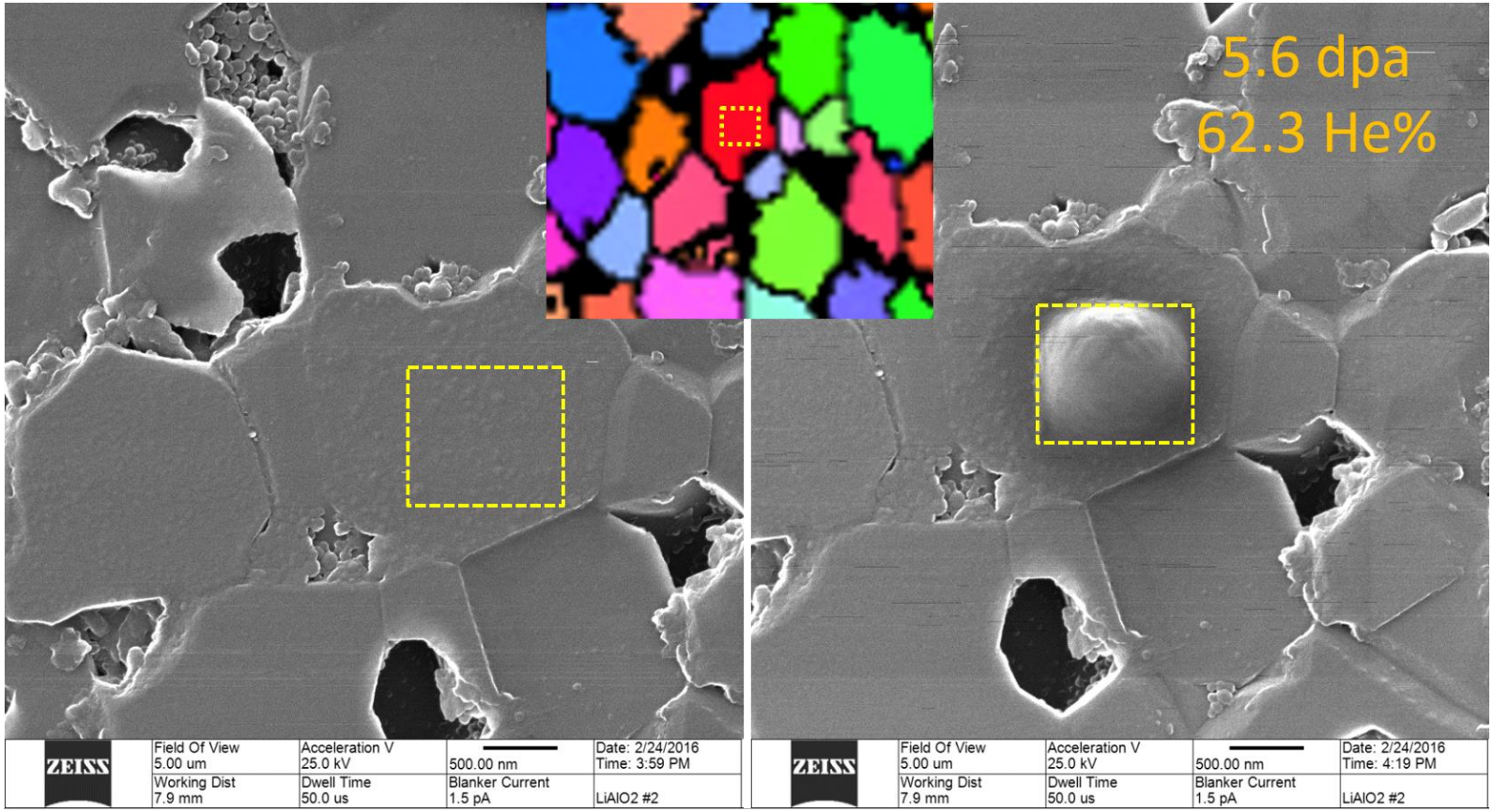
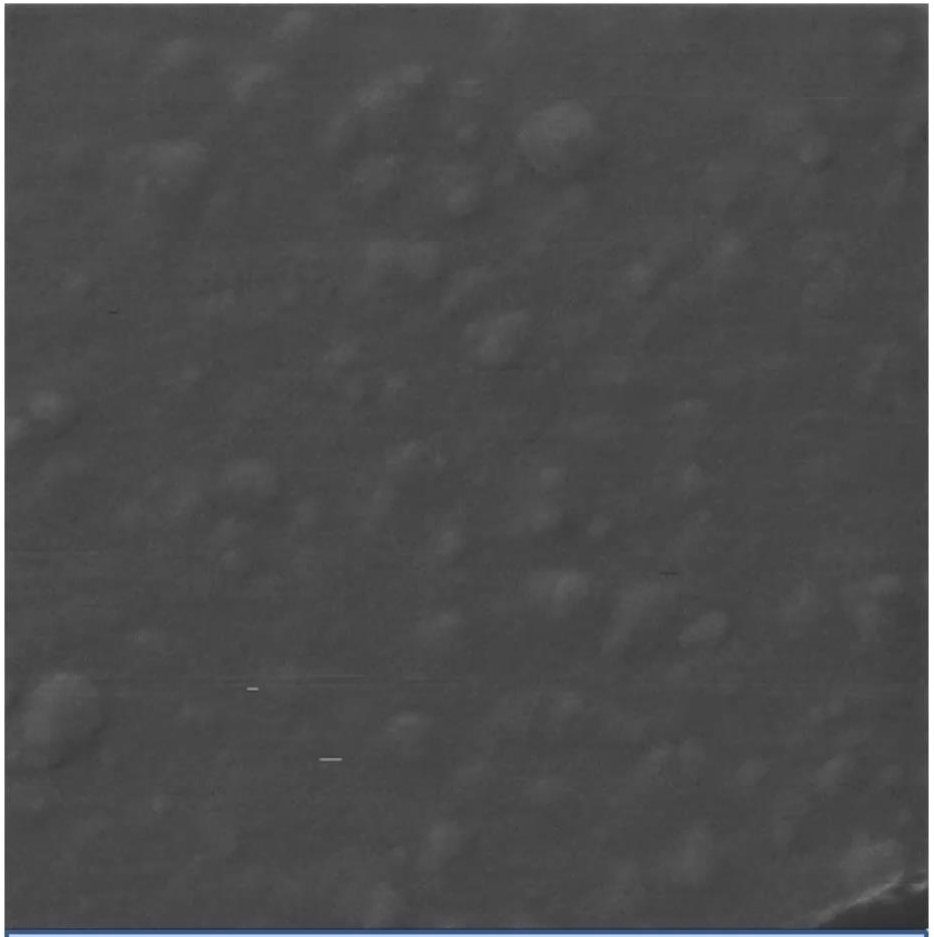
γ-$LiAlO_2$ irradiated with 25 keV He^+ at RT under HIM

(He⁺ ion projected range: 236 nm; max. 62.3 at.% He)

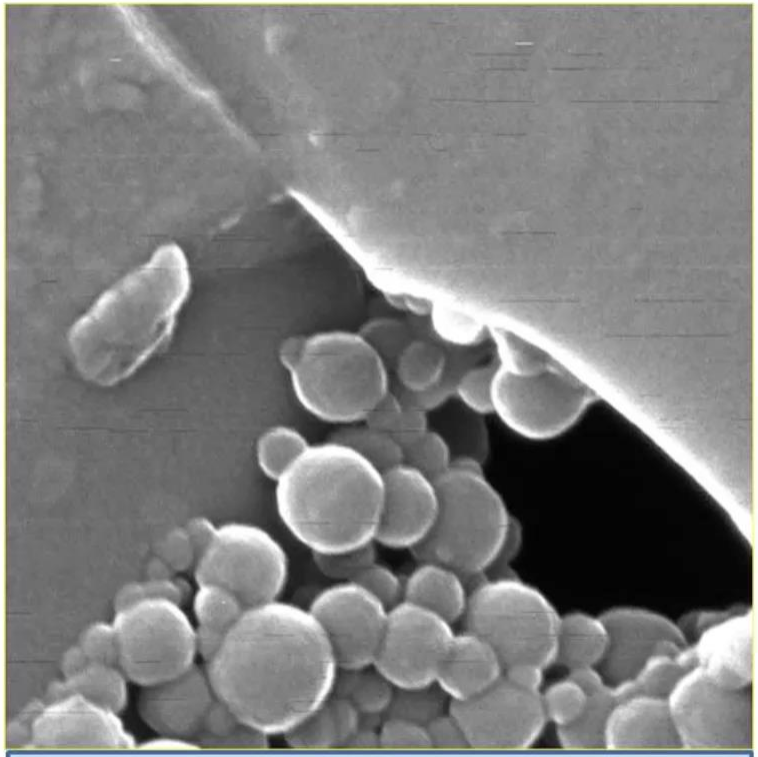


He Bubble Formation in a γ-$LiAlO_2$ Grain under HIM



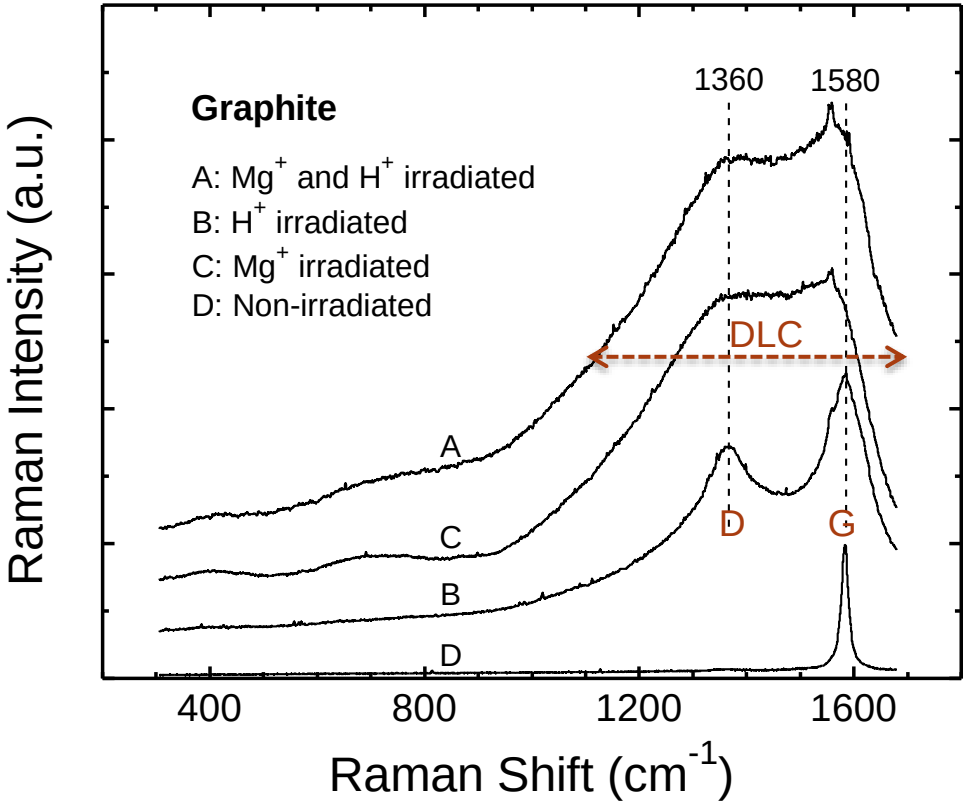
1.0 μm × 1.0 μm; 2.1 at.% He

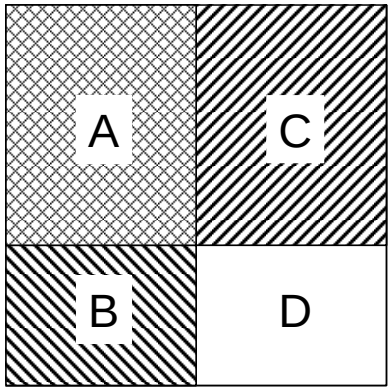
Microstructural Evolution of Amorphous SiO_2 Nanoparticles and $LiAlO_2$ at a Void under HIM



0.785 μm × 0.785 μm; 0.19 dpa

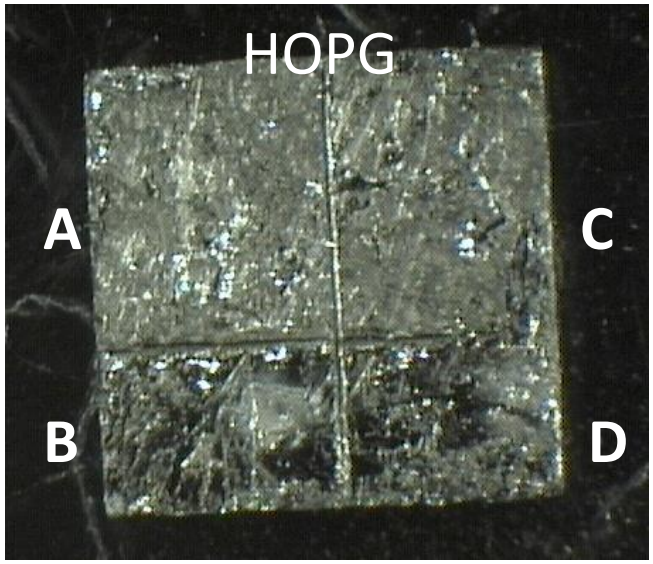
Mg^+ and H^+ Irradiated HOPG





keV	Mg^+/cm^2
70	7.1×10^{16}
120	1.0×10^{17}
200	2.0×10^{17}

0.78 MeV H^+
10 µm Al foil
1.05×10^{17} H^+/cm^2



G: Graphite peak at 1580 cm^{-1}
D: Disorder peak at 1360 cm^{-1}
DLC: Broad diamond like carbon peak ranging from 1100 to 1700 cm^{-1}