TEM Study of Tracks in Quartz Generated by Au Ion Irradiation

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Mineral Detection of Dark Matter (MDDM) Mini-school & Workshop

January 13~ 17, 2025, Blacksburg, VA

Michigan Ion Beam Laboratory

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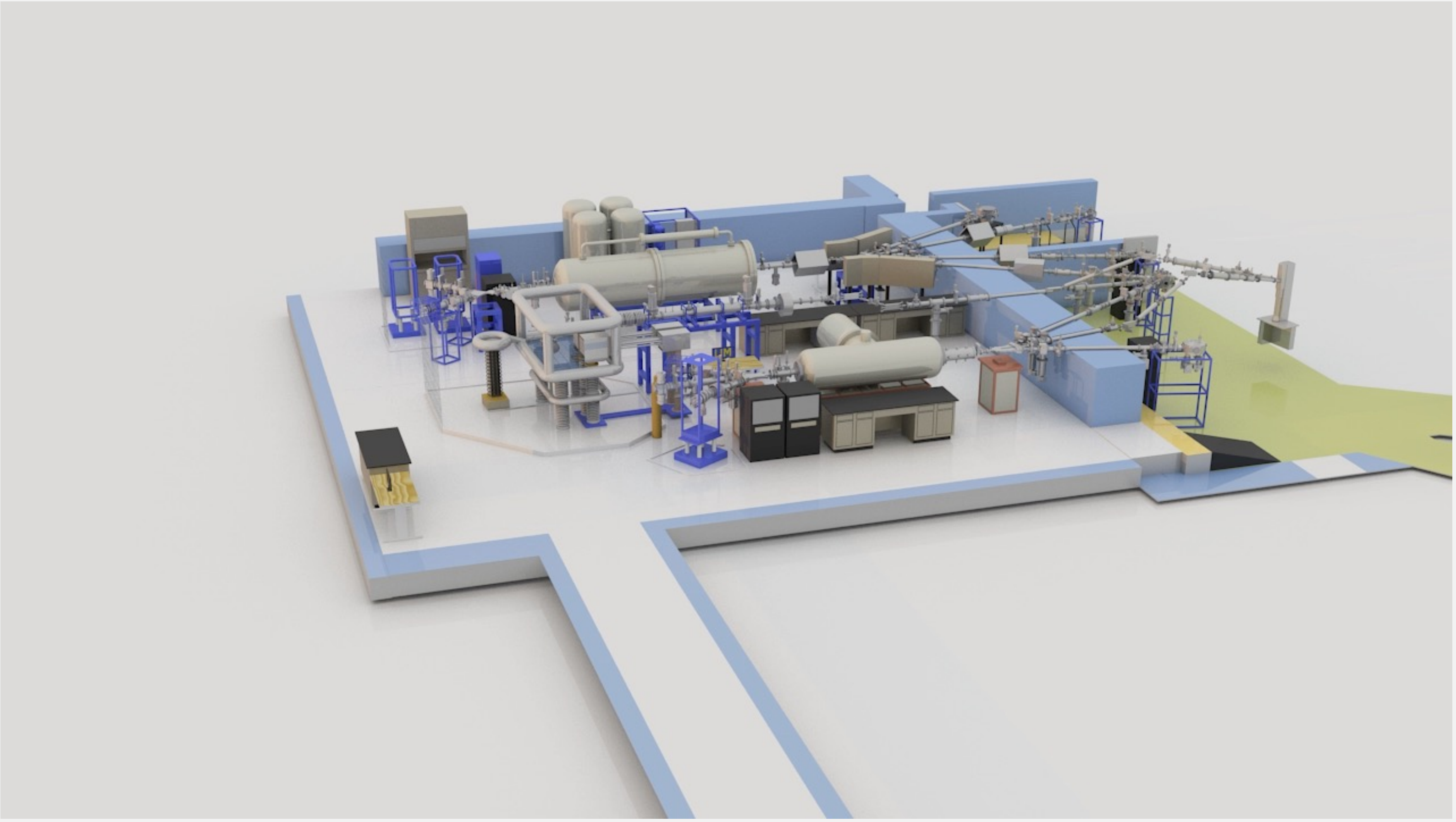
Outline

- Facilities in MIBL and $(MC)^2$ @ Umich
- Preliminary data from an Au irradiated quartz experiment
- Summary and future-plan

Facilities in MIBL @ Umich



Facilities in Michigan Ion Beam Laboratory (MIBL)

Accelerators
- 3 MV Tandem (Pelletron) (Wolverine)
- 1.7 MV Tandem (Tandetron) (Maize)
- 0.4 MV implanter (Blue)

Ion sources
- TORVIS (protons) – Wolverine
- SC SNICS, Peabody (sputter) – Wolverine
- Duoplasmatron, ECR (gases, eg He) – Maize
- Multi-cathode SNICS (sputter) – Maize
- Danfysik, multi-mode source – Blue

9 Beam lines

5 Target chambers
- ion irradiation
- irradiation accelerated corrosion
- multi-beam chamber
- 2 ion beam analysis chambers

300 kV FEI TEM*
- Dual beam interface for simultaneous damage and gas injection

Target temperature range: 77K to 1500K
Damage rate: < 10^{-5} dpa/s (protons), 10^{-3} dpa/s (heavy ions)
Irradiated area: up to 200 mm^2

Facilities in $(MC)^2$ @ Umich

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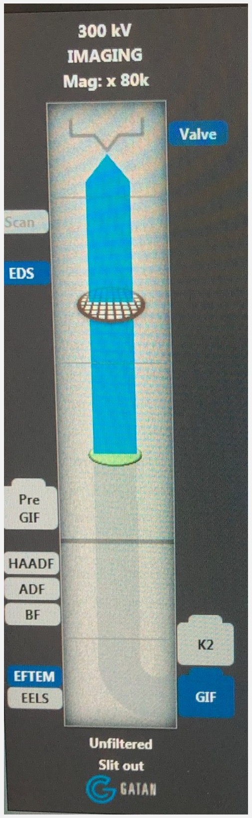
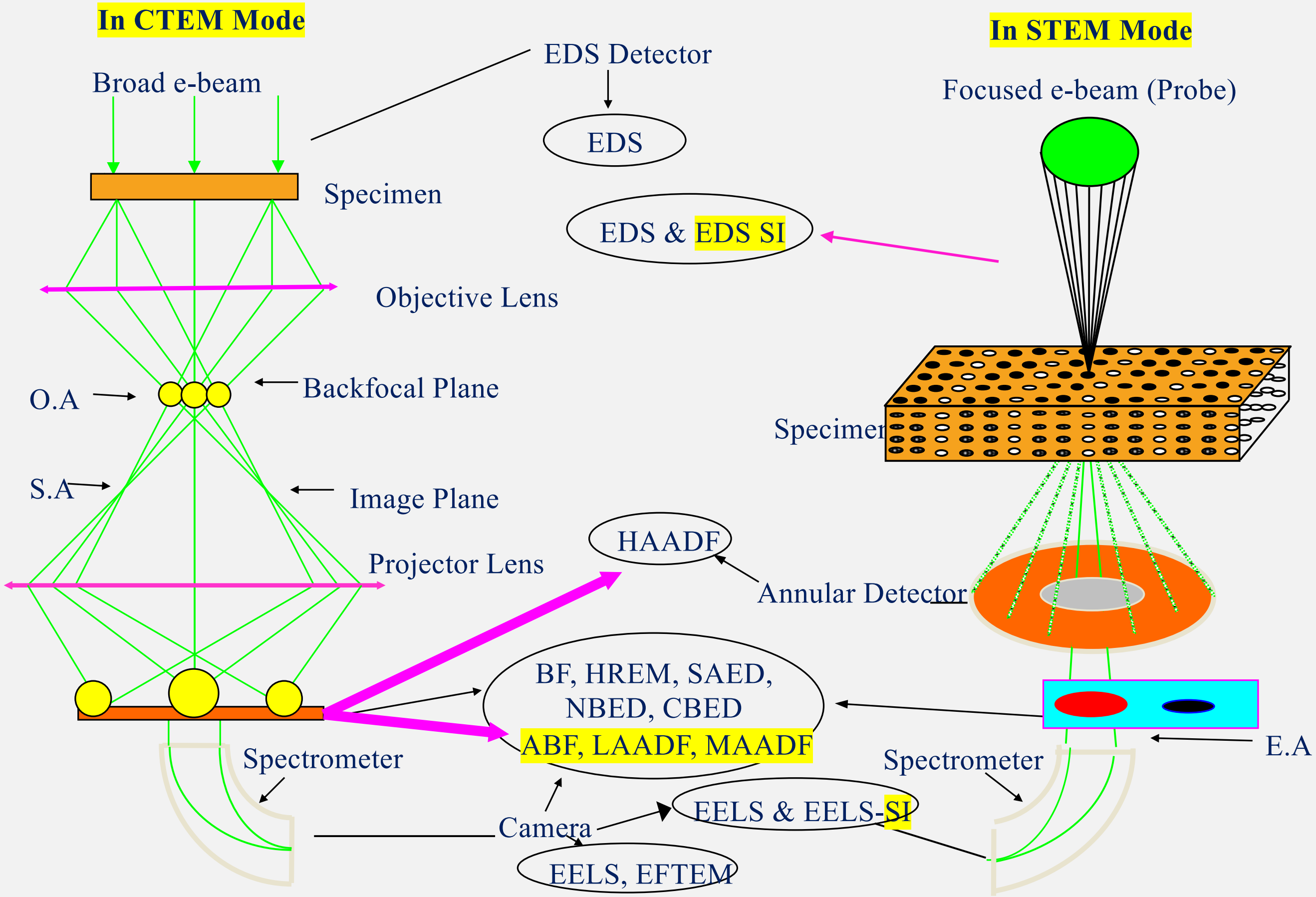
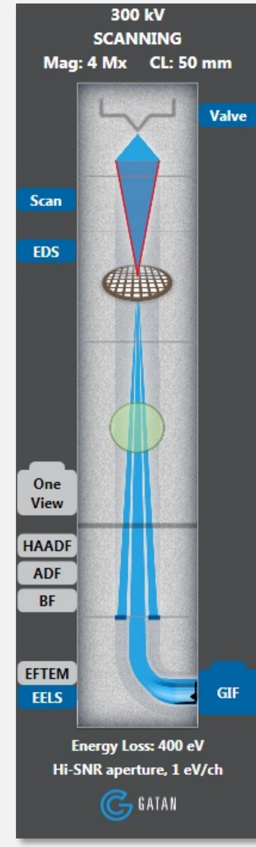
Why TEM?---A Variety of Techniques







- **Imaging**
- **Diffraction**
- **Spectroscopy**

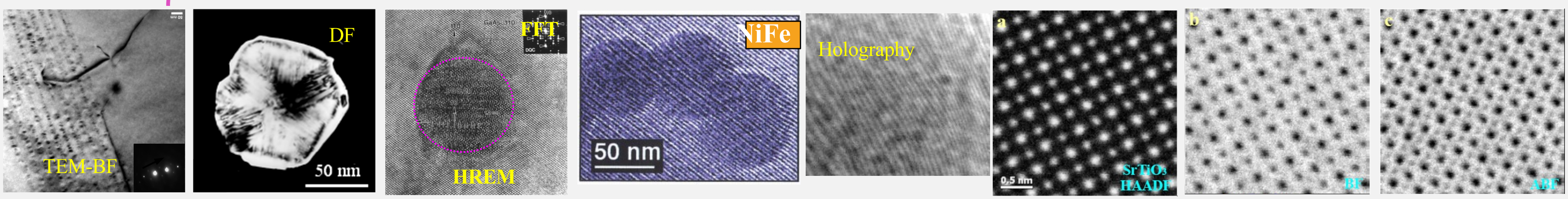
Special Operations: 4D-STEM/electron ptychography/fluctuation electron microscopy, DPC/iDPC, Electron holography, Lorentz electron microscopy, Electron magnetic circular dichroism (EMCD), and scanning near-field electron microscopy (SNEM), etc.

Why TEM?--- A Variety of Techniques: Examples

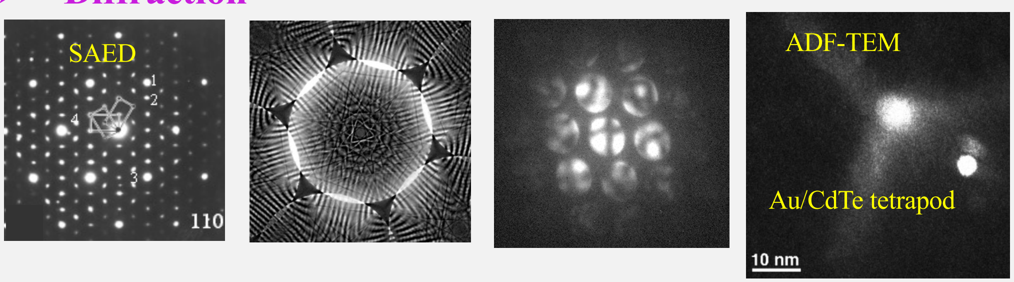
➢ Imaging

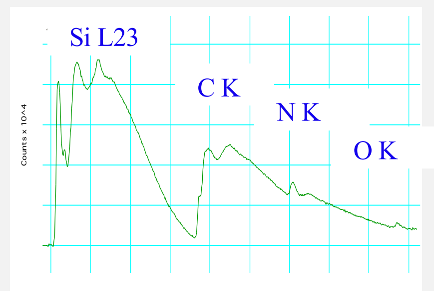
CTEM mode

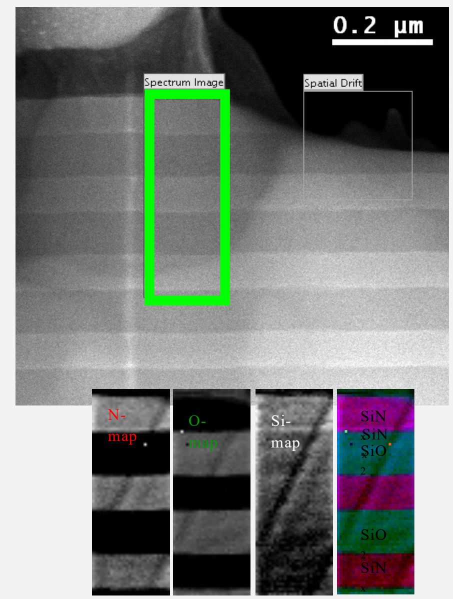
STEM mode



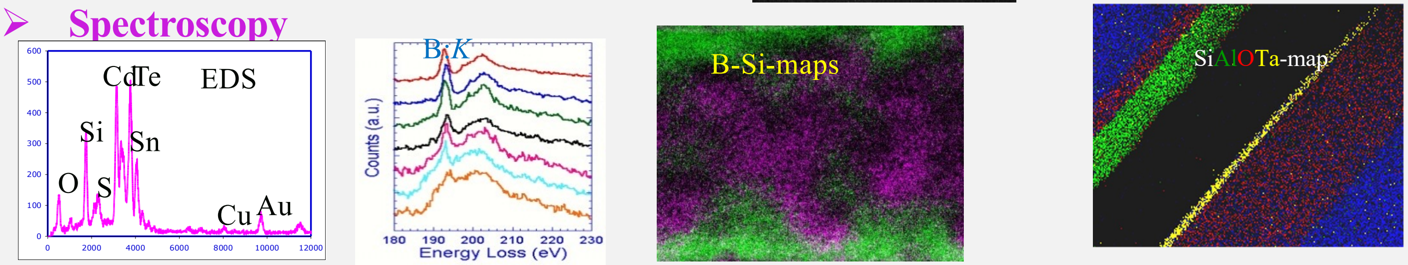
➢ Diffraction



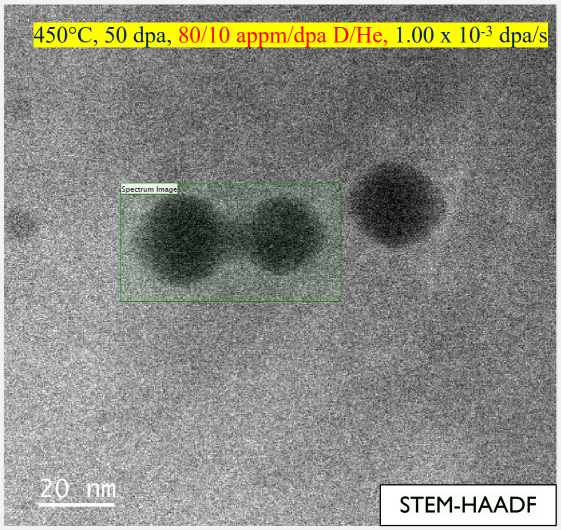


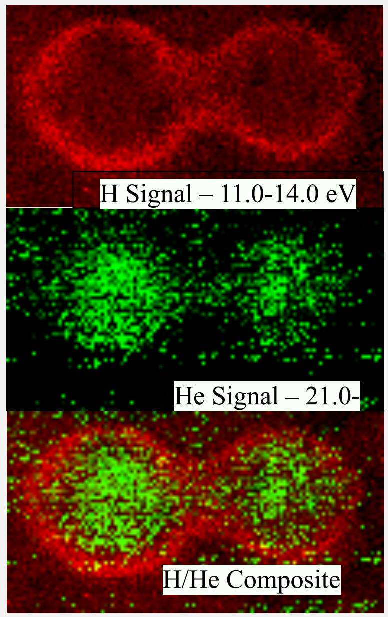


➢ Spectroscopy



Why TEM?---Imaging Light Elements By EELS Element Mapping

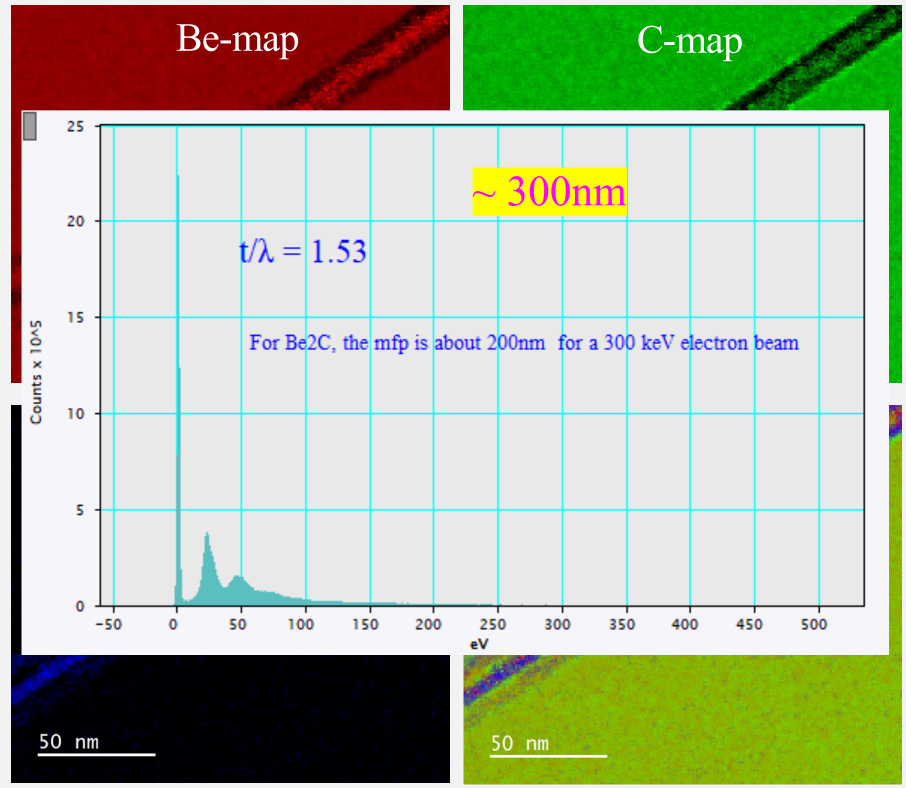




Logan Clowers et al.,

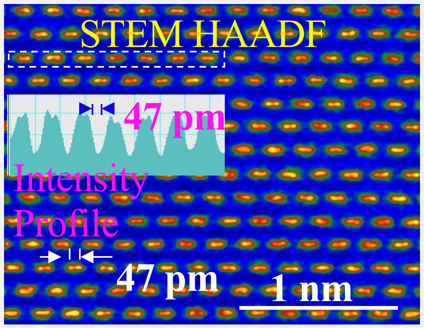
He/H gas bubbles in a Fe/He/H ions irradiated Stainless-Steel

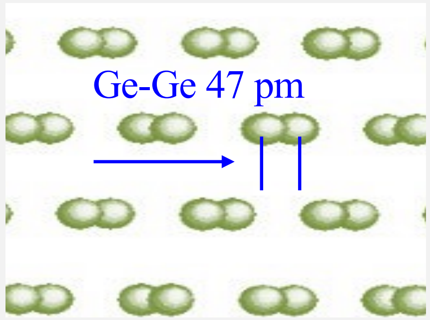
EELS data were acquired by Kai Sun using the JEOL-JEM3100R05 STEM/TEM in $(MC)^2$ @ Umich for Logan Clower from NERS @Umich

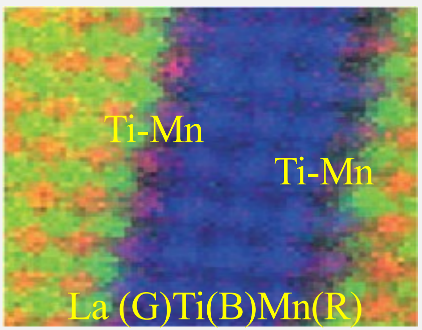


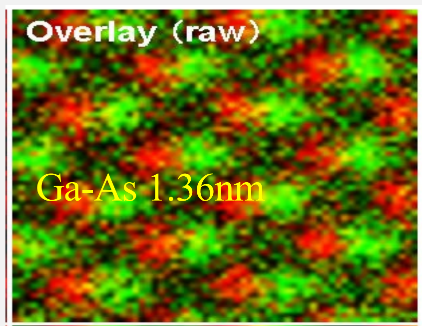
Be_2C sample from Diego Muzquiz from NERS@Umich

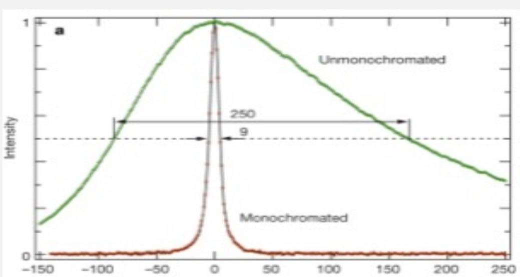
The Power of a Modern TEM









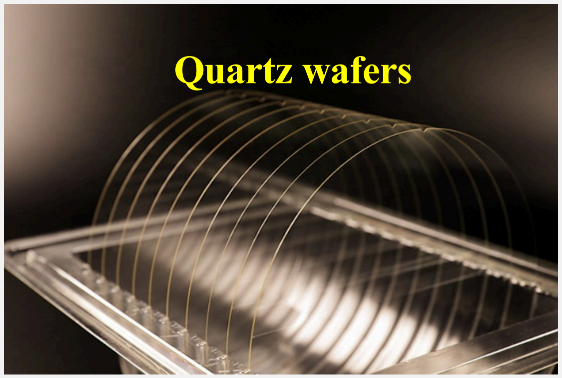


9 meV wide FWHM monochromated ZLP compared to the energy distribution of an unmonochromated beam produced by the system's cold field emission electron gun (~4meV)---Phonon scattering regime.

OL Krivanek *et al.* *Nature* **514**, 209-212 (2014)

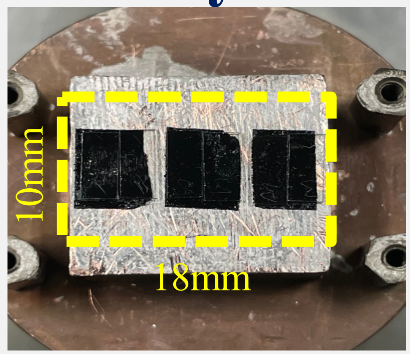
- **Resolution jumps to sub-Å: Increased measurement precision**
  - Sub-Angstrom spatial resolution imaging
  - Atomic-scale chemical imaging-Ultrafast STEM SI System and Large EDS detectors
- **Energy resolution for EELS has reached ~4 meV.**
- **In-situ Cs-TEM study: Pole-piece gap can be larger --- good for in-situ study**
- **Gentle TEM study (~ 15 keV still reached atomic scale resolution)**
- **3D tomography and Stereo-vision 3D imaging**
- **4D microscopy: ~ fs to ns temporal resolution.**
- **Scanning Confocal Electron Microscopy (SCEM)**
- **Electron Ptychography**
- **More**

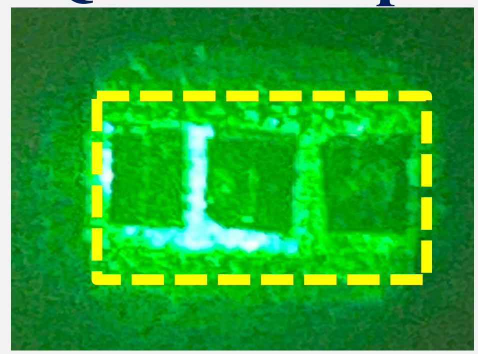
Creation of Ion Tracks in Synthetic Quartz: Experimental

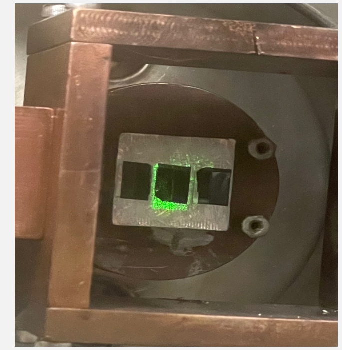


- Hydrothermally synthesized High purity (optical grade) single crystal quartz wafers from Vritra Technologies
- 5inch x 0.5mm double-side polished

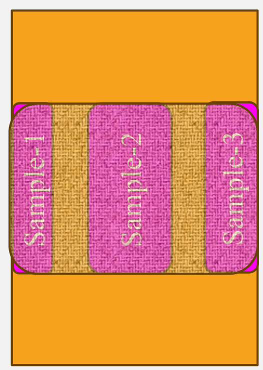
- **Ion beam parameters**: 15 MeV Au^{5+} ion beam with current of 5nA (~ 6.25 x 10^9 Au^{5+}/s and a 180mm^2 raster area (~ a fluence of 3.47 x $10^9 Au^{5+}/cm^2s$) at room temperature.
- Three groups of samples (~2mm x5mm) were irradiated for one experiment with the samples loaded on the ion beam stage illustrated below:



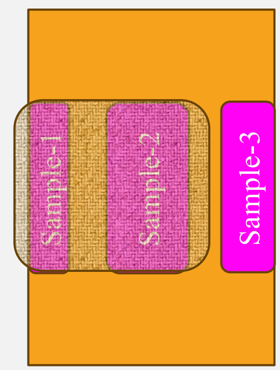




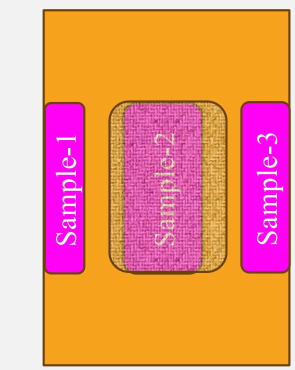
Samples on the ion beam stage



Sample-3: 5.21 x $10^{10} Au^{5+}/cm^2$



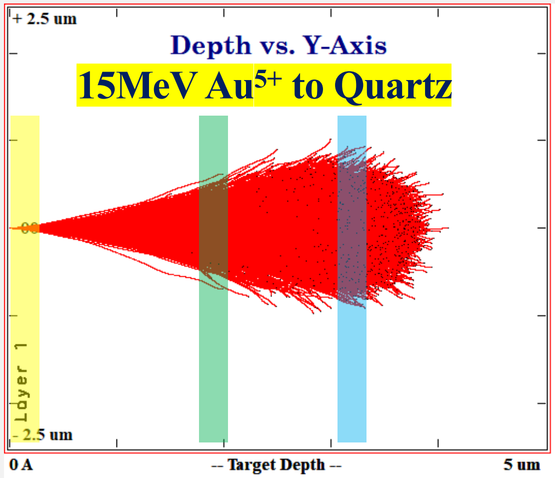
Sample-1: 1.04 x $10^{11} Au^{5+}/cm^2$

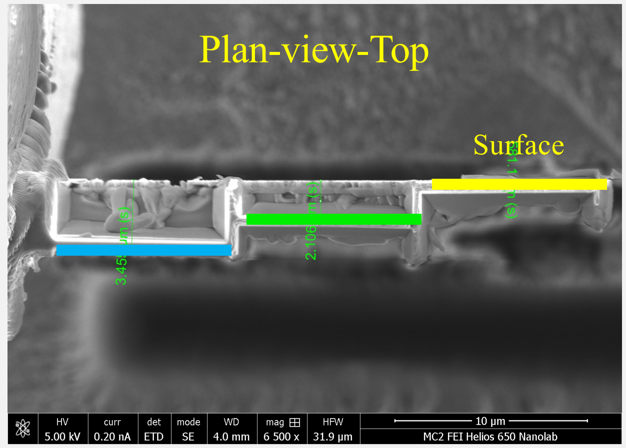


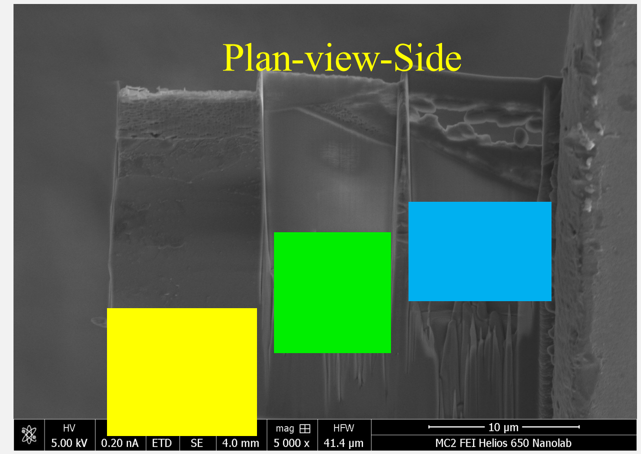
Sample-2: 1.56 x $10^{11} Au^{5+}/cm^2$

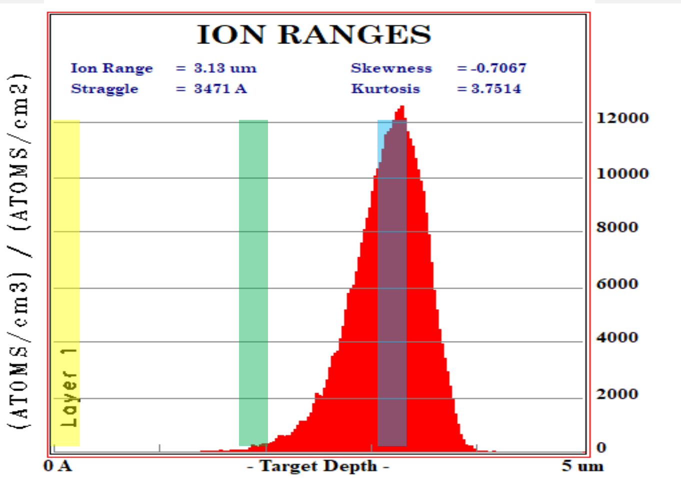
- **Time was controlled using iPhone clock with roughly ~1s error!**
- **To reach the ~10^6 ions/cm^2, only 1 μs is needed for such a beam current.**

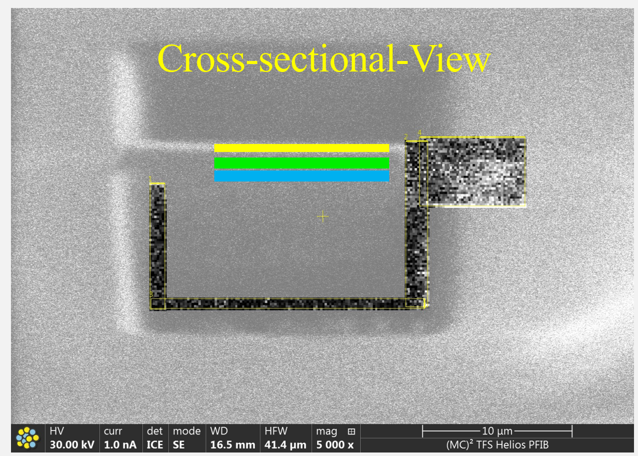
FIB Sampling Based on SRIM Simulations



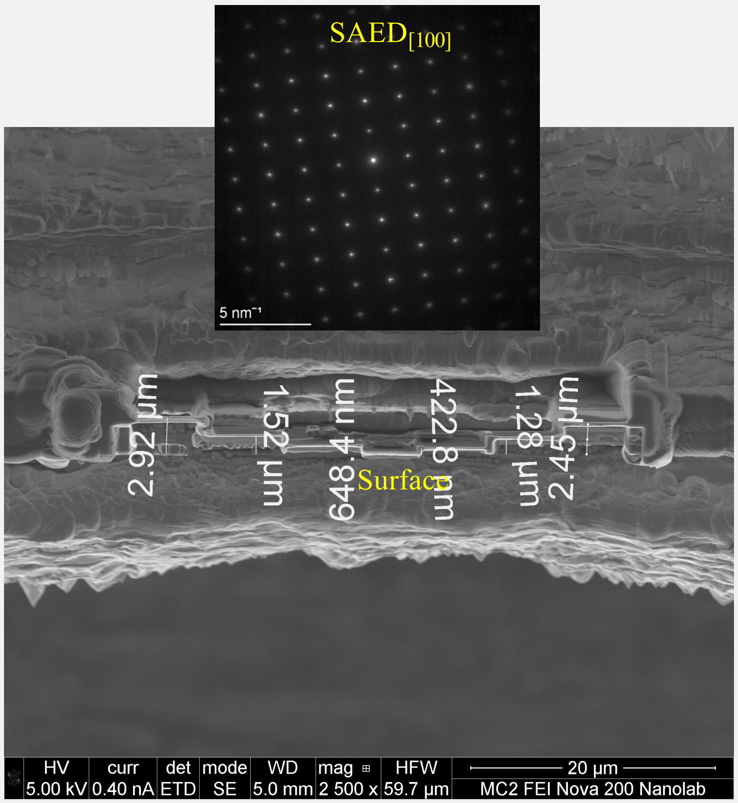






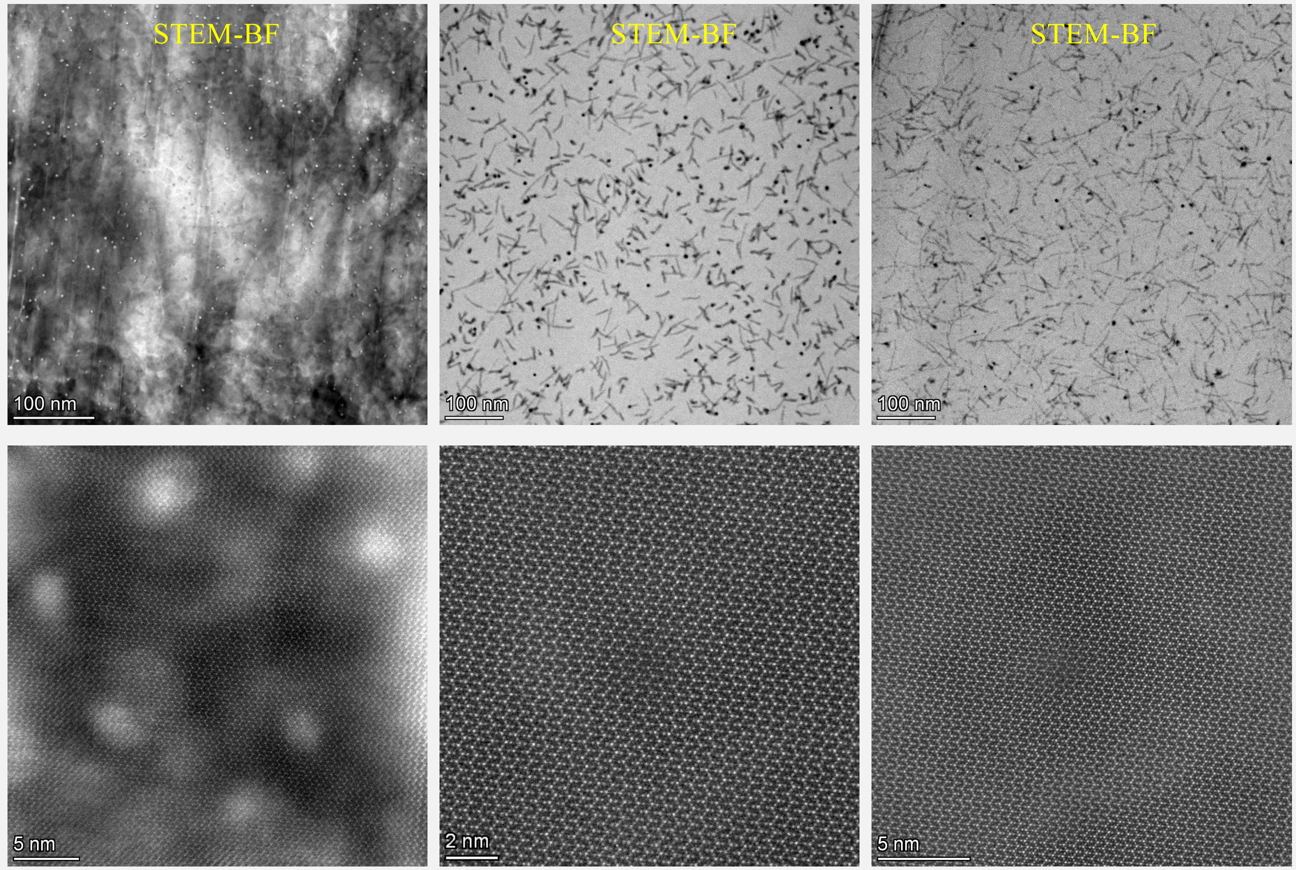


“Pseudo-3D” Imaging of Ion Tracks in Nature Olivine



15MeV to Olivine: Sample-2: $1.74 \times 10^{11} Au^{5+}/cm^2$

FIB sectioning vs. AFM + etching



Surface layer | ~500nm depth | ~2 μm depth

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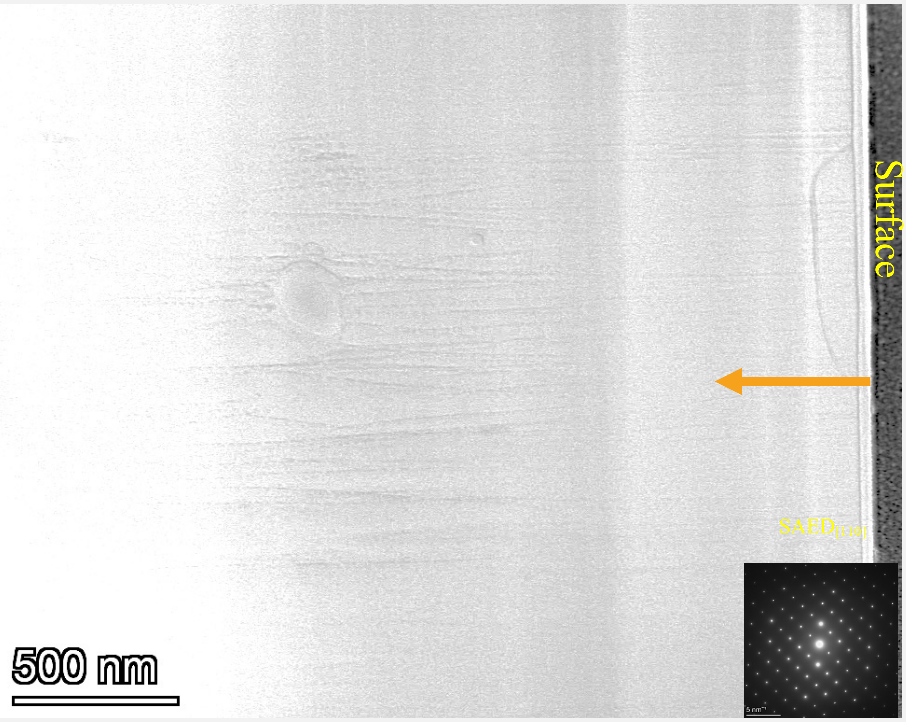
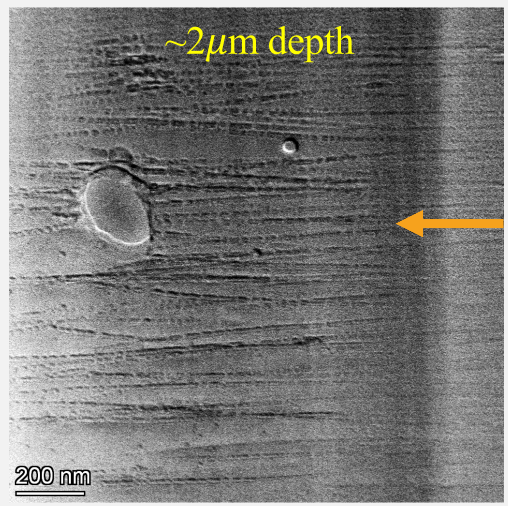
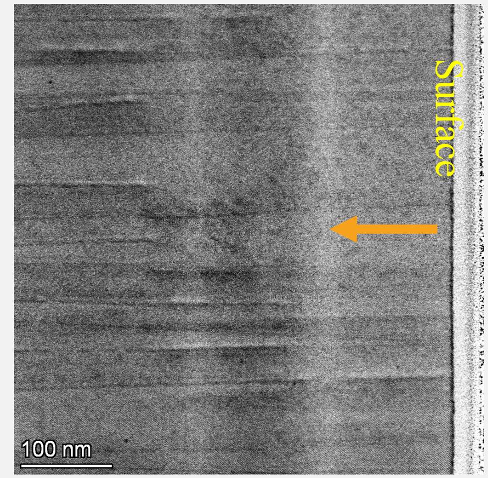
X-STEM Imaging of Au Ion Tracks in Synthetic Quartz



Sample-1: 1.04 x 10^{11} Au^{5+}/cm^2

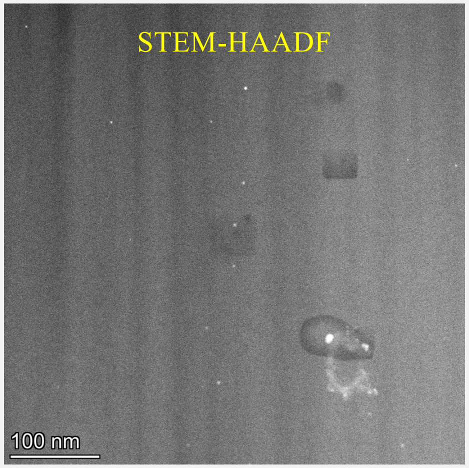
Au^{5+} ion beam direction

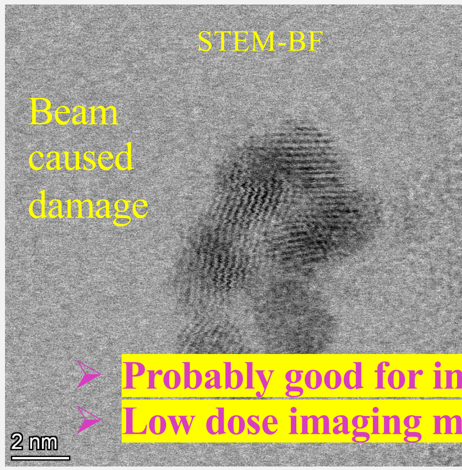


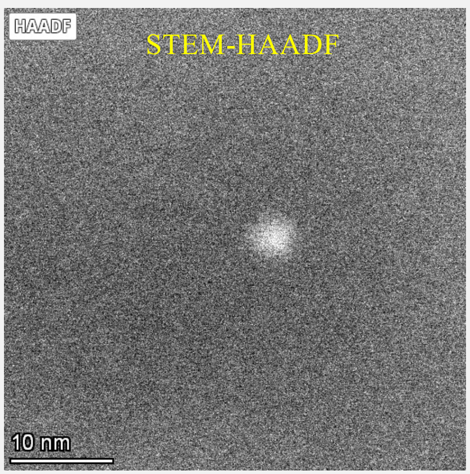


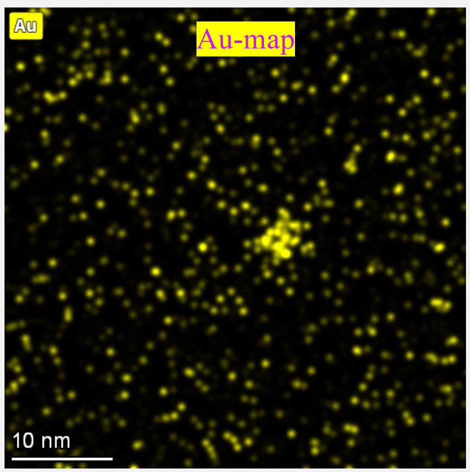
STEM-BF Images

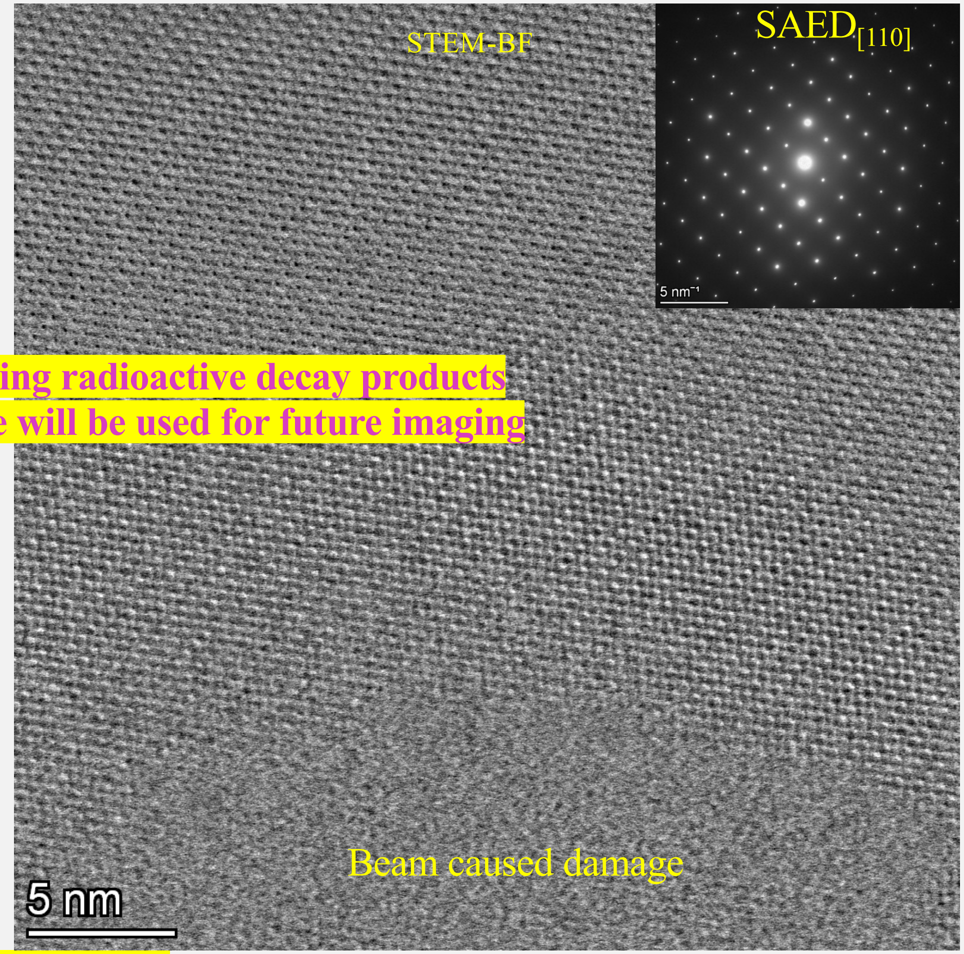
X-STEM Imaging of Au Atoms in Synthetic Quartz







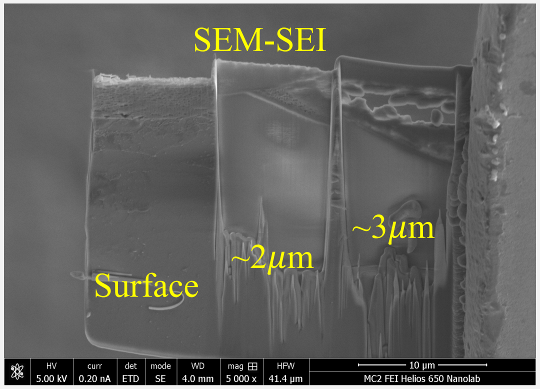


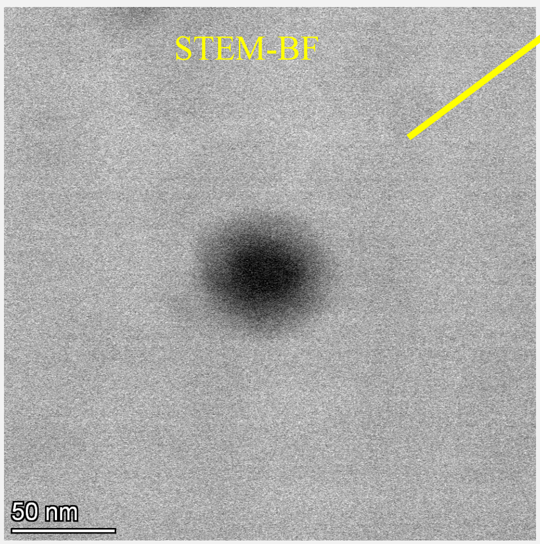


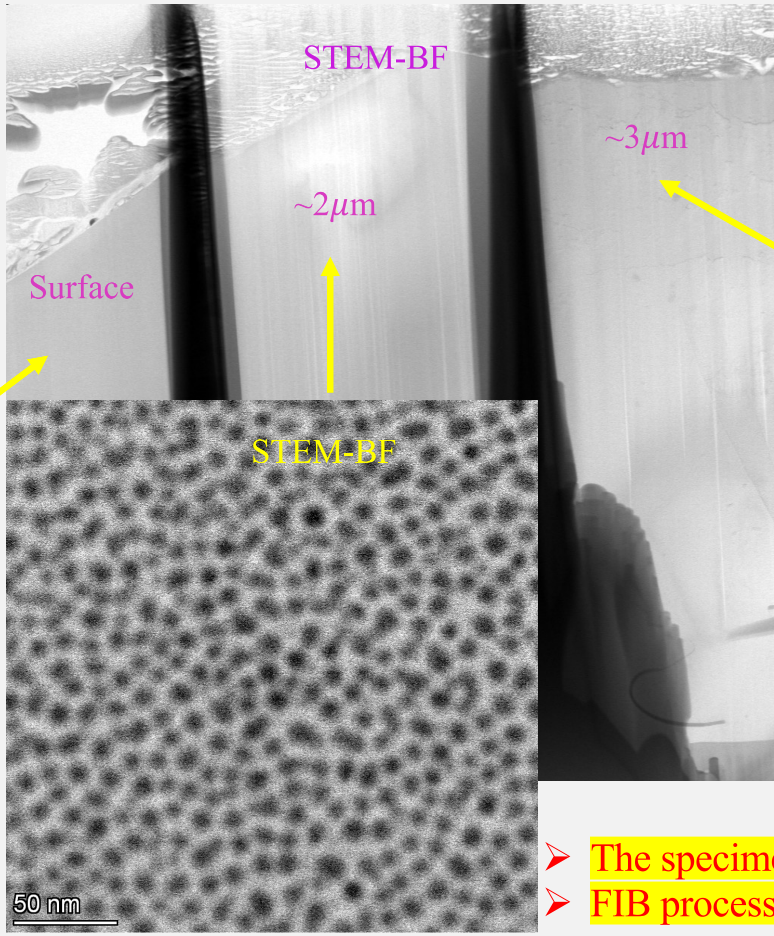
- **Probably good for imaging radioactive decay products**
- **Low dose imaging mode will be used for future imaging**

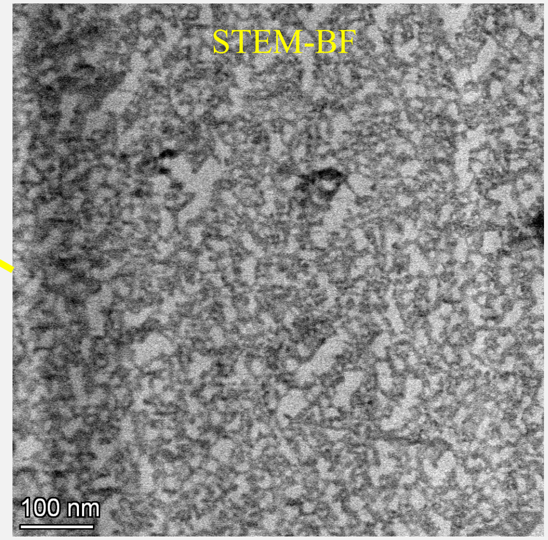
Sample-1: 1.04 x 10^{11} Au^{5+}/cm^2

"Pseudo-3D" Imaging of Ion Tracks in Synthetic Quartz









Sample-1: 1.04 x 10^{11} Au^{5+}/cm^2

- The specimen has been badly damaged during FIB!
- FIB process needs to be optimized for future work.

New Techniques to Be Used: Cathodoluminescence (CL)

REZA EBADI *et al.* PHYS. REV. D **104,** 015041 (2021)

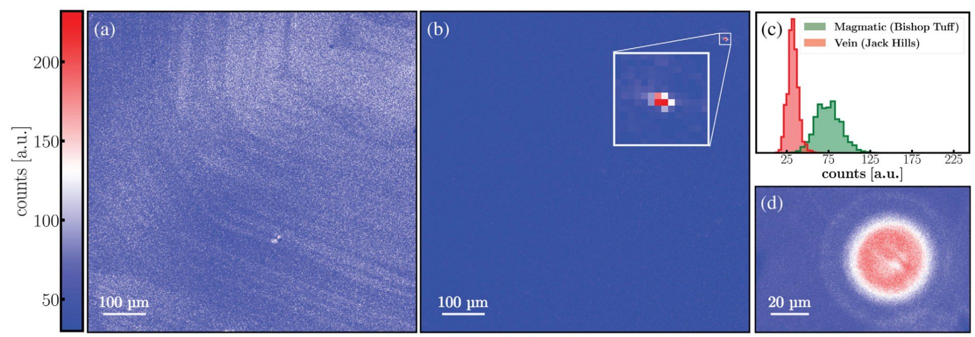
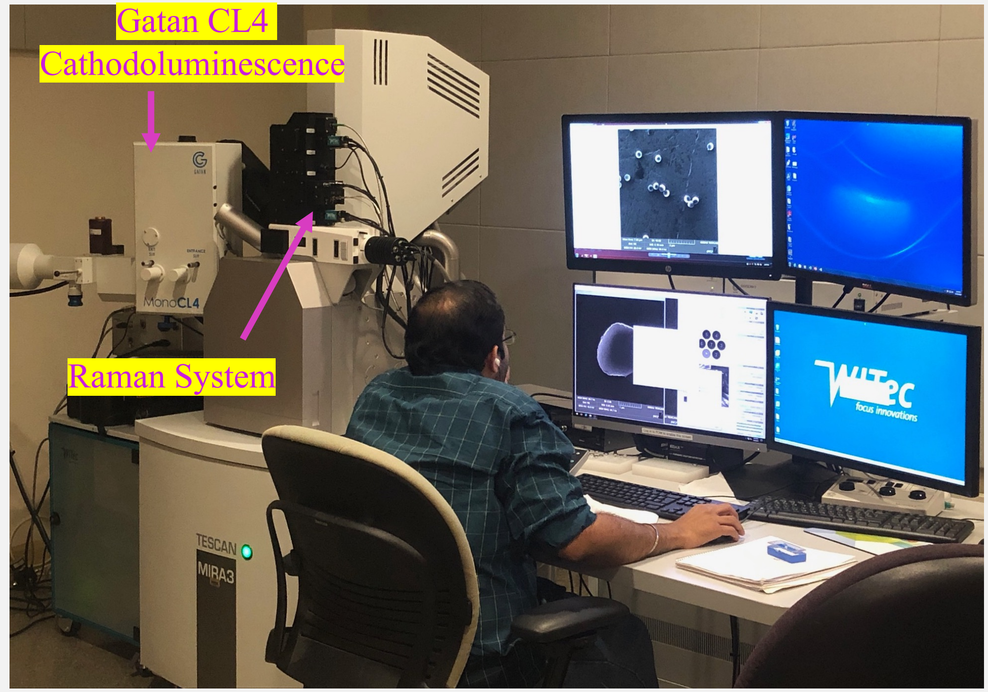


FIG. 2. Example quartz sample characterization. SEM-CL images of two samples, (a) magmatic quartz from Bishop Tuff with Ti concentration 51 ± 6 ppm, and (b) vein quartz from Jack Hills with Ti concentration 5.2 ± 6.5 ppm, measured on a mass spectrometer. The scan rate is 20 s/mm^2 with 1.5 μm resolution for magmatic quartz and 5 s/mm^2 with 3 μm resolution for vein quartz (we forecast the full-scale UHDM experiment time and resources using these values). In (b) we identify a few high-count pixels in the vein quartz image, which demonstrates the possibility of high-resolution detection of concentrated CL emission. The inset shows a zoomed-in image of the region of interest with high-count pixels. These pixels could be a melting track intersection, which needs to be investigated by correlating multiple sections as described in the text. (c) Normalized histogram of the pixel counts in arbitrary units for each of the two sample SEM-CL images. Vein quartz shows a lower CL noise level as well as smaller variation, making it a suitable target for our detection proposal. (d) SEM-CL signal from a uranium halo [measured in a different quartz sample from those shown in (a) and (b)]. Microscopic uranium inclusions have decayed over time; the fission products from these inclusions create crystal lattice damage, which emits CL upon excitation by the SEM. The CL signal from a UHDM particle track would also result from crystal lattice defects at and around the track of melted quartz. Any such uranium halos in a UHDM search would be disqualified as potential damage tracks by lack of correlated damage in other slices of the sample.

Reza Ebadi et al., Ultraheavy dark matter search with electron microscopy of geological quartz, PHYSICAL REVIEW D 104, 015041 (2021).



TeScan RISE SEM/Raman/CL in $(MC)^2$ @ Umich

3 μm resolution is probably good for well isolated track imaging

New Techniques to Be Used: X-ray Microscopy

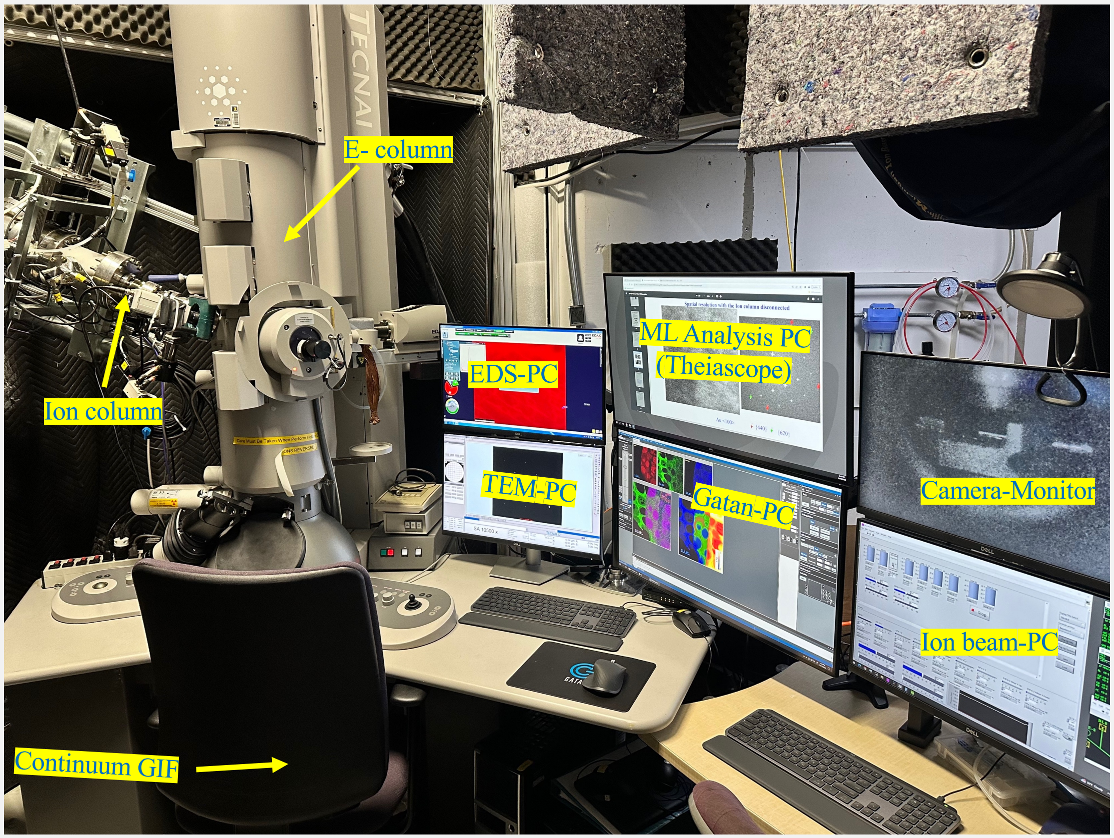
ZEISS Crossbeam
FIB-SEM for High Throughput 3D Analysis

- Combine imaging and analytical performance of a high-resolution FE-SEM) with the processing ability of a next-generation focused ion beam (FIB) and a Laser Beam included.
- Performance:
  - Maximize your SEM insights
  - Increase your FIB sample throughput
  - Experience best 3D resolution in your FIB-SEM analysis
- Coming this Month
- Good for large sample cutting

ZEISS Xradia 810 Ultra
Nanoscale X-ray Imaging

- Nanoscale 3D X-ray imaging at a spatial resolution down to 50 nm and 16 nm voxel sizes
- 3D and 4D *in situ* experiments
- Quantification of nanostructures and using the data for modelling input. --- To be coming next Month

In-Situ Ion Irradiation TEM-Tecnai F30: One of the Two in the US

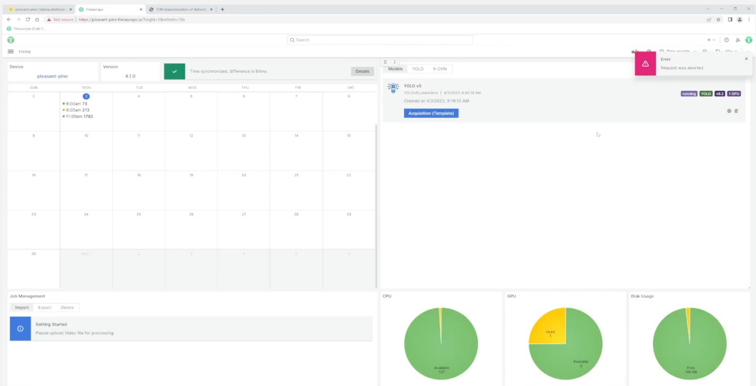


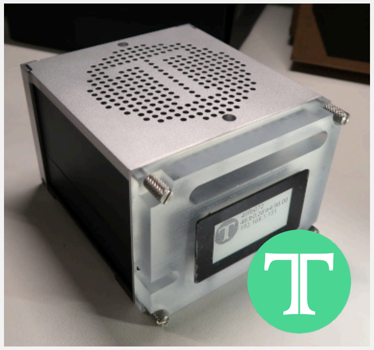
- **TEM Specs**---TF30 TEM/STEM
- **Gun**: Thermo-FEG, HT: 100, 200 and 300keV
- **Operation modes**: CTEM: ~0.1 nm lattice resolution and STEM: 0.34nm point-to-point resolution
- **Cameras**: a 2Kx2K UltraScan Pre-GIF CCD and a 2Kx2K GIF CCD
- **Detectors**: HAADF, ADF and BF
- **EDS**: EDAX Elite 70mm^2 SDD detector.
- **Gatan energy filter system (GIF)**
- **Different Holders: DT-regular and heating holders**
- **Remote control**
- **Theiascope™ platform** for in-situ dislocation loops/precipitates analysis
- **SenseAI system** for fast STEM imaging (~10fps)
- **4D STEM**

Ion Beams --- with Two beam lines linked

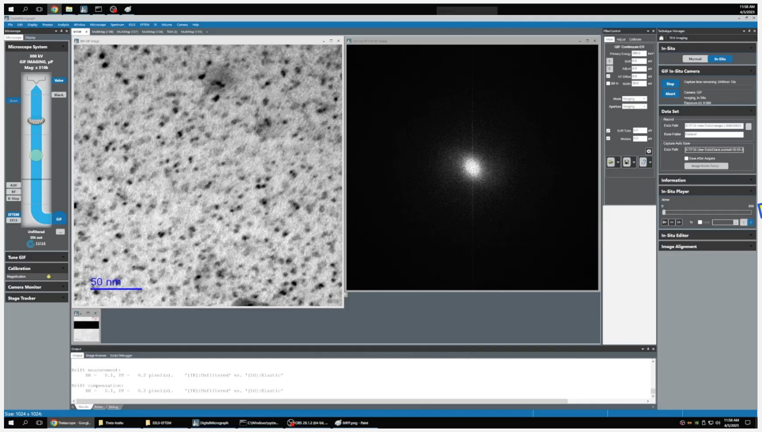
- A 400kV NEC accelerator equipped with a Danfysik Model 921 ion source can provide a wide range of ions (H+ to Au) ions with energy 20keV ~1.6MeV
- An Alphatross NEC ion source can deliver H and He ions in the energy range 5~30keV
- Dual beam irradiation capability
- Remote control

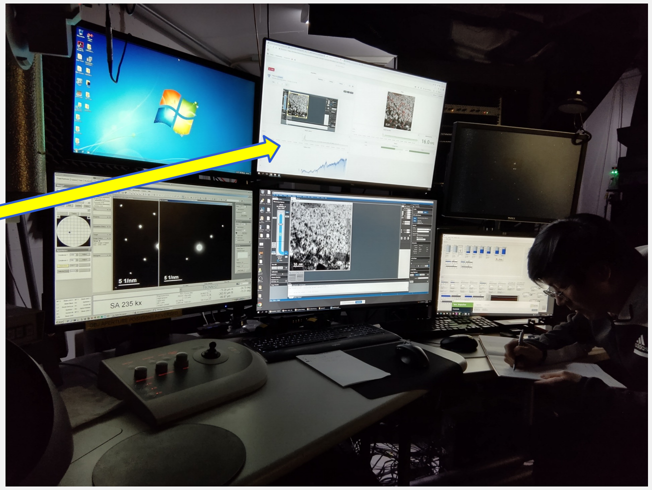
ML System: Theiascope™ System for In-situ Data Analysis





*C.R. Field & K.G. Field, U.S. Patent Application No. 17/718,805



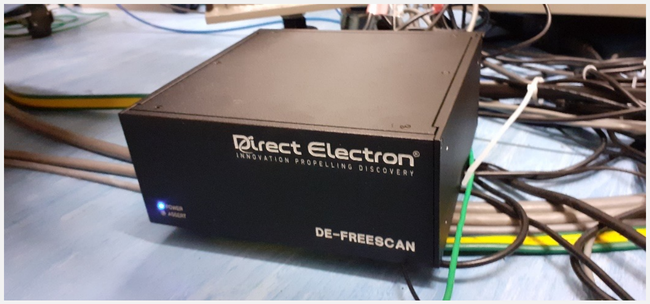


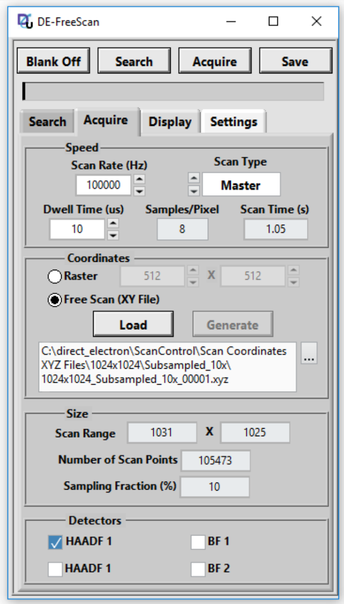
Courtesy H. Li (UM) and Theia Scientific, LLC

- Real-time quantification of irradiation induced Dislocation loops/voids during an in-situ TEM ion irradiation exposure.
- I am thinking to quantify abundant track data using this machine learning system.

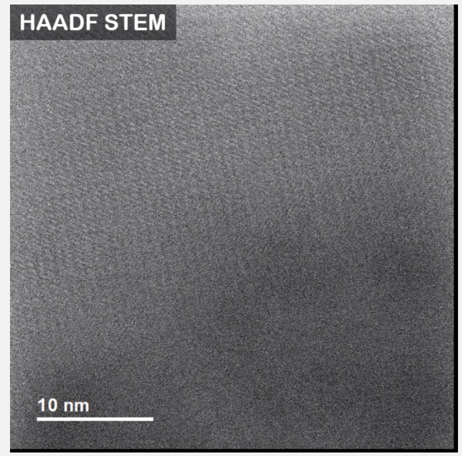
Sense-AI for Fast STEM Imaging

Modification to the Microscope





$Rb_3NdP_2O_8$- a very e-beam sensitive ionic compound



10% Random Sampling

Reconstruction

Sub-sampling scan controller plugs into external control and uses pre-generated masks. Images reconstructed currently using Matlab scripts. ~ 10fps scanning rate with good image quality

Courtesy SenseAI and University of Liverpool

➢ Possible for imaging ion tricks in electron beam sensitive samples

Summary and Future Plan

- ❑ **Summary**

- ➢ High energy Au ions irradiation has been used for generation of ion tracks in a synthetic quartz single crystal sample.
- ➢ STEM has been used for imaging Au ion tracks in the synthetic quartz.

- ❑ **Things to do next**:

1) Perform O ion irradiation (self-ion irradiation) in to both synthetic and natural quartzes with smaller fluences.
2) Optimize FIB parameters for better TEM specimens and image tracks under low dose modes for both synthetic and natural quartzes (before and after irradiation).
3) Try other techniques like XRM and CL.
4) Introduce AI and ML systems for data collection and analysis