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Radiation Damage In Accelerator Target Environments

Porosity evolution and crystal damage in proton irradiated POCO graphite

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MATERIALS RESEARCH FACILITY

2023 RaDIATE Collaboration Meeting
Radiation Damage In Accelerator Target Environments
Hosted by Brookhaven National Laboratory
June 26–30, 2023

University of BRISTOL Fermilab EMAM RaDIATE Collaboration
Radiation Damage In Accelerator Target Environments

Self-introduction

- Mechanical engineering (MEng) and Nuclear Science and Engineering (MSci) from University of Bristol, UK.
- PhD in University of Bristol, co-funded by UK. Engineering & Physical Sciences Research Council (EPSRC) and US. Fermilab.
- PhD thesis title: *Microstructural and property evolution of nuclear graphite under extreme conditions*

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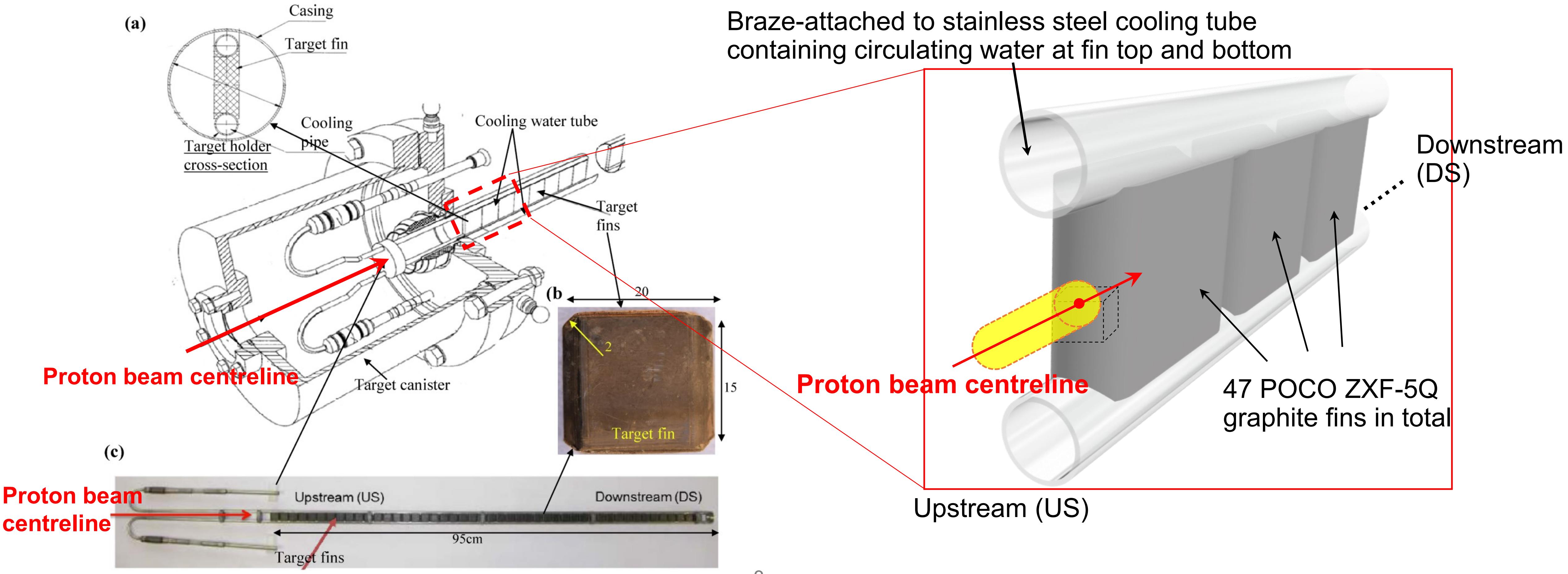
Outline

- Introduction
- Materials
- Experimental methods
  - Focused ion beam-scanning electron microscopy tomography (FIB-SEM tomography)
  - Raman spectroscopy
  - Measurement schematics
- Part I: Porosity evolution studied by FIB-SEM tomography
  - Results
  - Conclusion
- Part II: Crystal damage studied by Raman spectroscopy
  - Results
  - Conclusion
- Future work

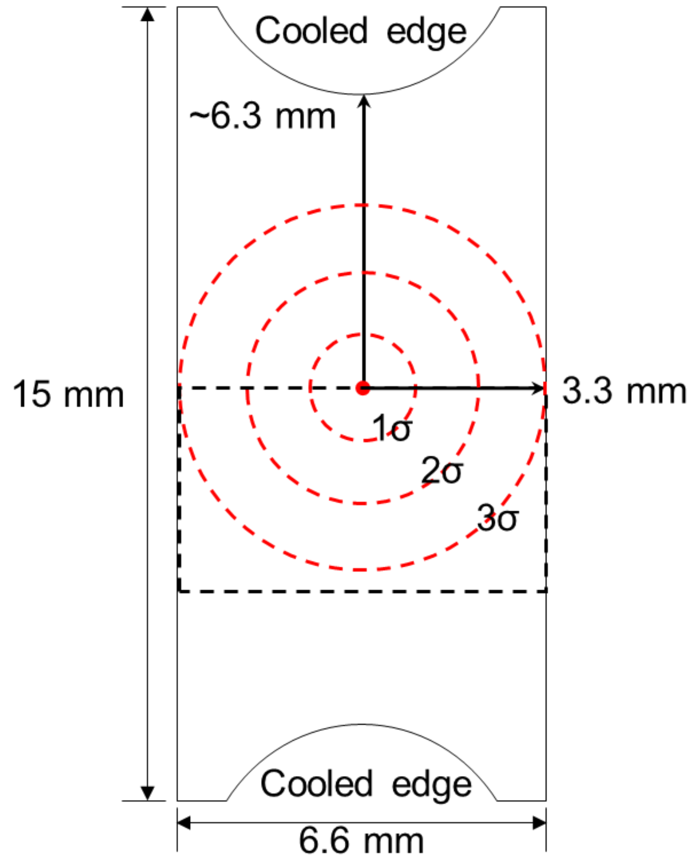
Introduction

An upstream (US) POCO ZXF-5Q graphite fin irradiated in the NT02 target system at NuMI beamline of Fermilab.



Introduction

A fractured upstream (US) POCO ZXF-5Q graphite fin irradiated in the NT02 target system at NuMI beamline of Fermilab.



Beam Gaussian 1σ radius = 1.1 mm

- 340 kW, 120 GeV proton beam, pulsed (cycling time 1.87s), and high intensity with a Gaussian beam profile (1σ radius = 1.1 mm).
- Rapid thermal cycling, target rose from ~60°C to ~370°C in 10 μs and cooled back to ~60°C before repeating.
- Temperature gradient from beam centre (~370 °C) to cooling tube edge (~60 °C).
- Inert non-circulating Helium environment (no oxidation).
- A gradient of irradiation damage was created due to both fluence and temperature distribution, with their exact distributions not resolved.
- It has not been possible to deconvolute fluence and temperature's contribution towards the total irradiation damage, among with many other factors.
- Porosity evolution and crystal irradiation damage were studied across these damage gradients.

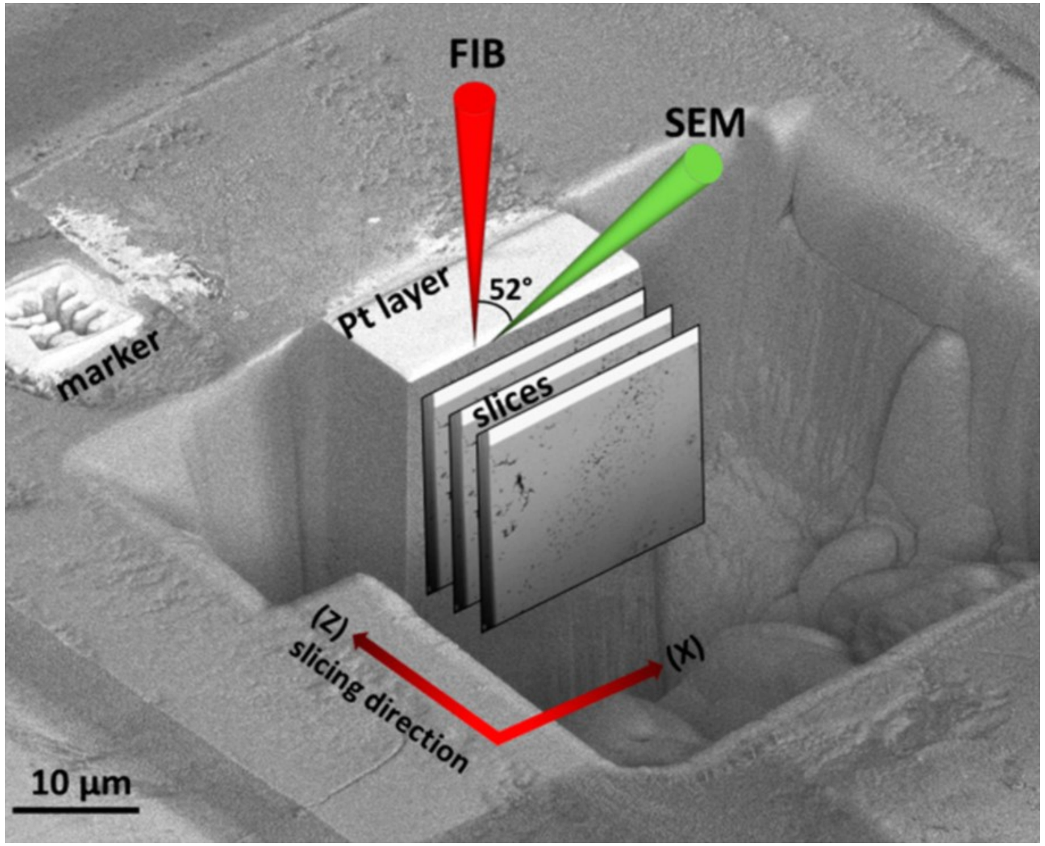
Materials

A piece of POCO ZXF-5Q graphite fragment irradiated in the NT02 target system at NuMI beamline of Fermilab.

Non-irradiated bulk properties	POCO ZXF-5Q [3]	IG 430 [3]	Gilsocarbon graphite [4]
Average grain size (μm)	1	10	500
Apparent density (g/cm^3)	1.78	1.82	1.81
Young's modulus (GPa)	14.5	10.8	10-11
Total porosity (vol.%)	20	14-21	20
Compressive strength (MPa)	175	90	70
Flexural strength (MPa)	112	54	25-28
Tensile strength (MPa)	79	37.2	~ 20
Graphitisation temperature (°C)	2500	≥ 2800	~ 2800
Thermal conductivity (W·m^{-1}·K^{-1})	70	140	138
Coefficient of thermal expansion (10^{-6} K^{-1})	8.1	4.8	4.7-4.9
Isotropic ratio	<1.03	1.09	1.04 (based on CTE ratio)

Experimental methods

Focused ion beam-scanning electron microscopy tomography (FIB-SEM tomography)

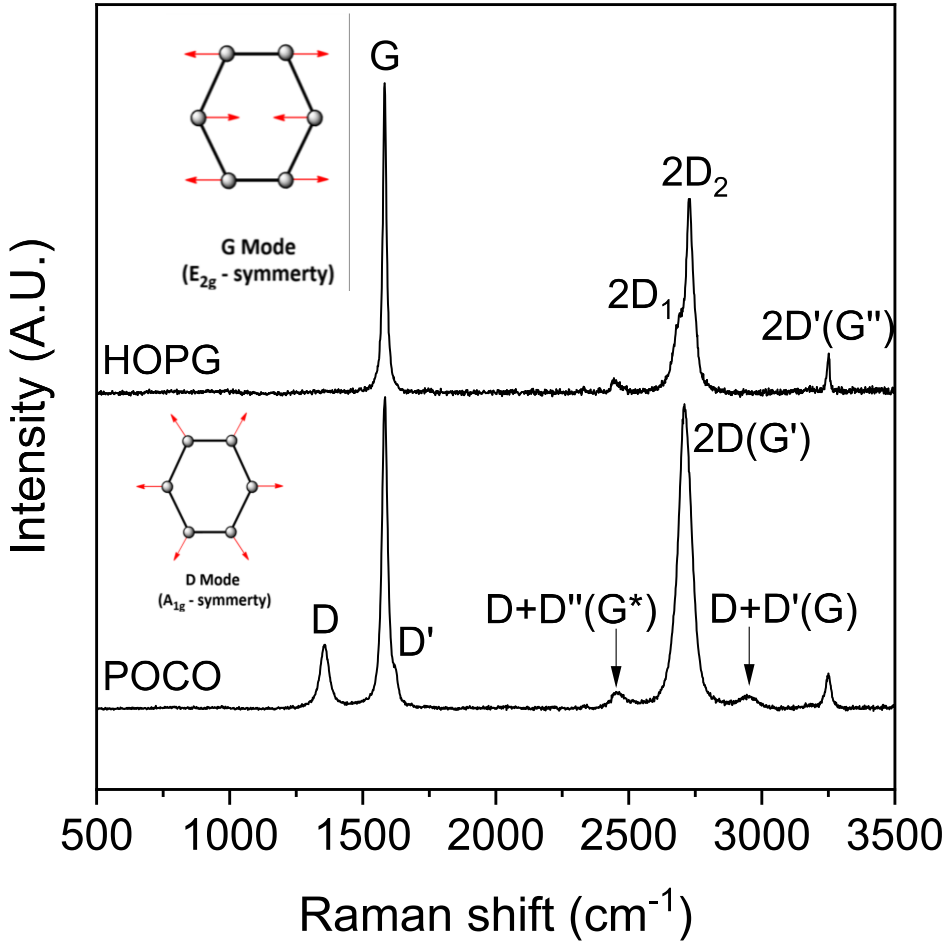


Simplified schematic of how FIB tomography works [5]

- FIB-SEM tomography is a destructive technique that physically mills through the material volume.
- Freshly milled material cross-section by ion beam is imaged by SEM.
- Alternatively repeating this process gives a stack of tomographic images that could be used for porosity segmentation and quantification.

Experimental methods

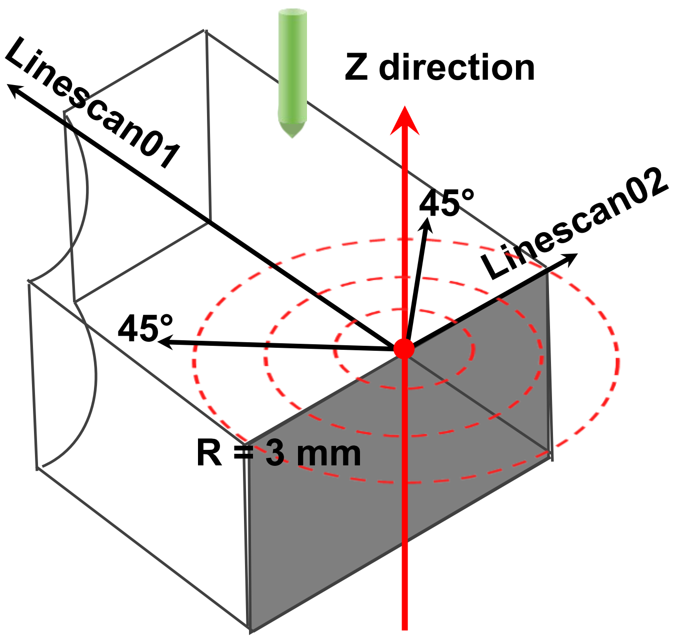
Raman spectroscopy



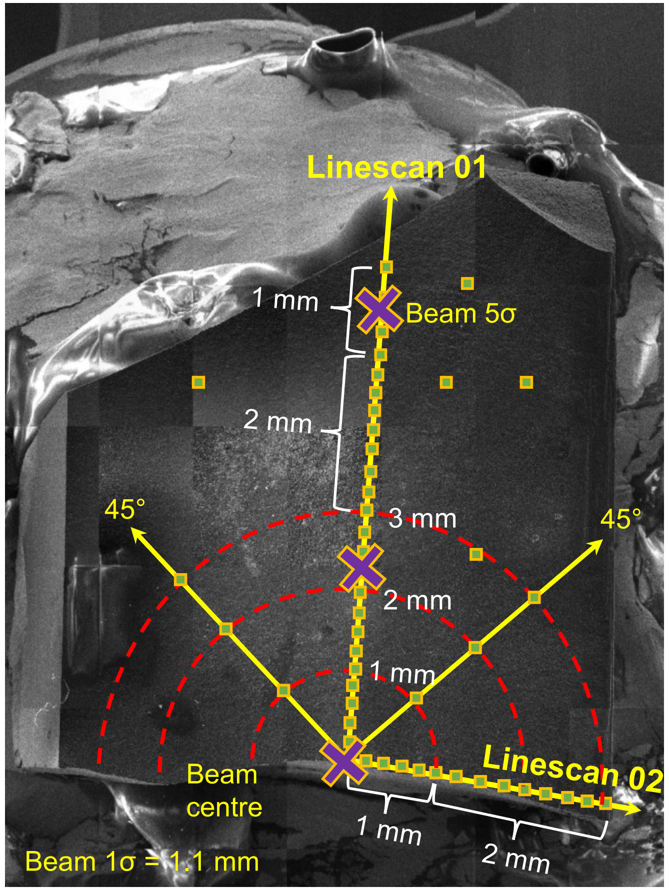
- Raman is surface-based laser technique.
- G-band is a common in graphitic materials, corresponding to sp^2 carbon network.
- D-band is an indicator of crystal damage level, A_{1g} symmetry of the in-plane breathing mode in aromatic rings.
- *I(D)/I(G)* ratio and G-band evolution (position and width) will provide rich information and be utilised in this work.

Experimental methods

Measurement schematics (a piece of fractured POCO graphite from beam upstream)



Schematic of the sample



Stitched SEM image showing top-down view

- **Raman mapping sites**
  48 maps across linecan01, linescan02 and 45° directions. Each map contains at least 100 points.

- **FIB-SEM tomography sites**
  Beam centre
  Beam 2σ
  Beam 5σ

Outline

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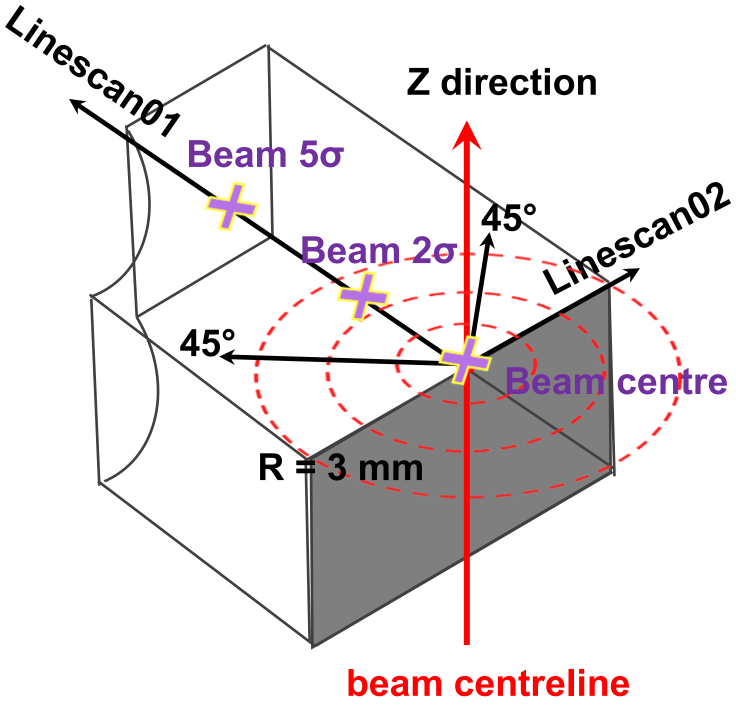
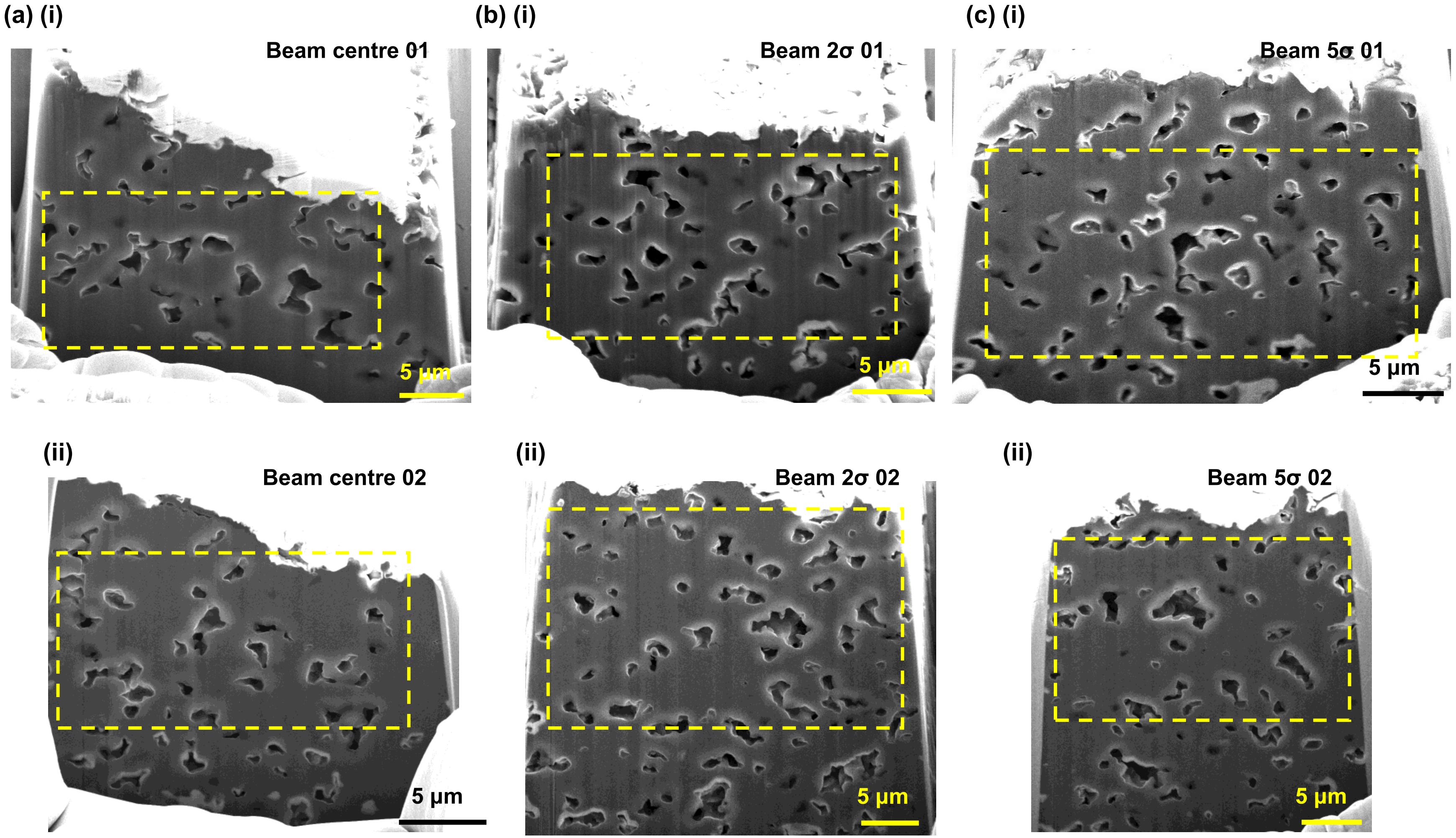
Part I: Porosity evolution studied by FIB-SEM tomography

Results: Example of SEM images



Schematic of the sample



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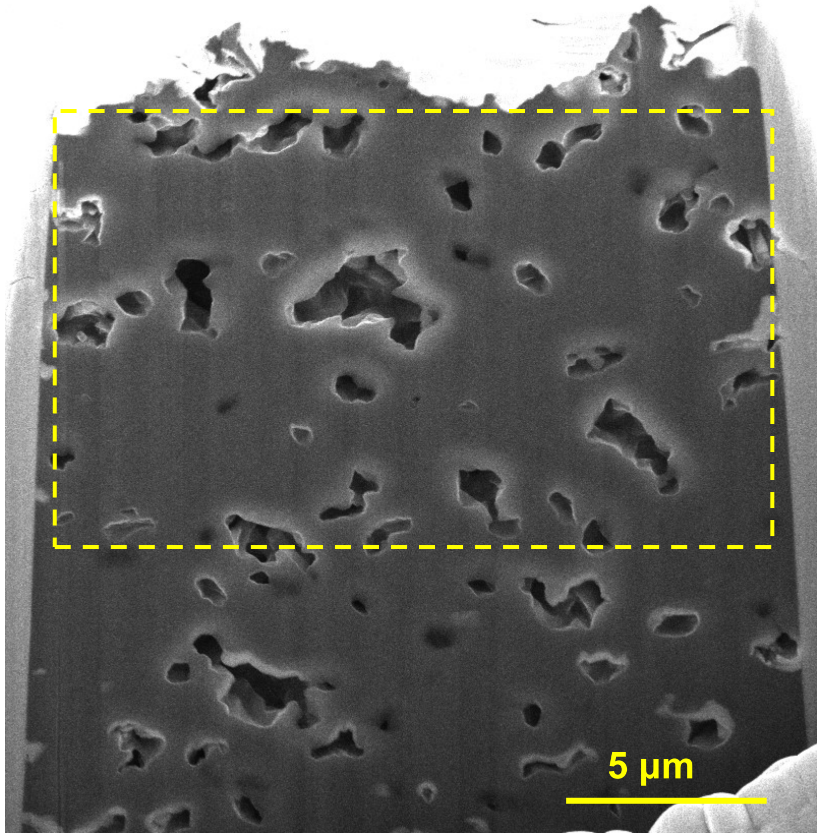
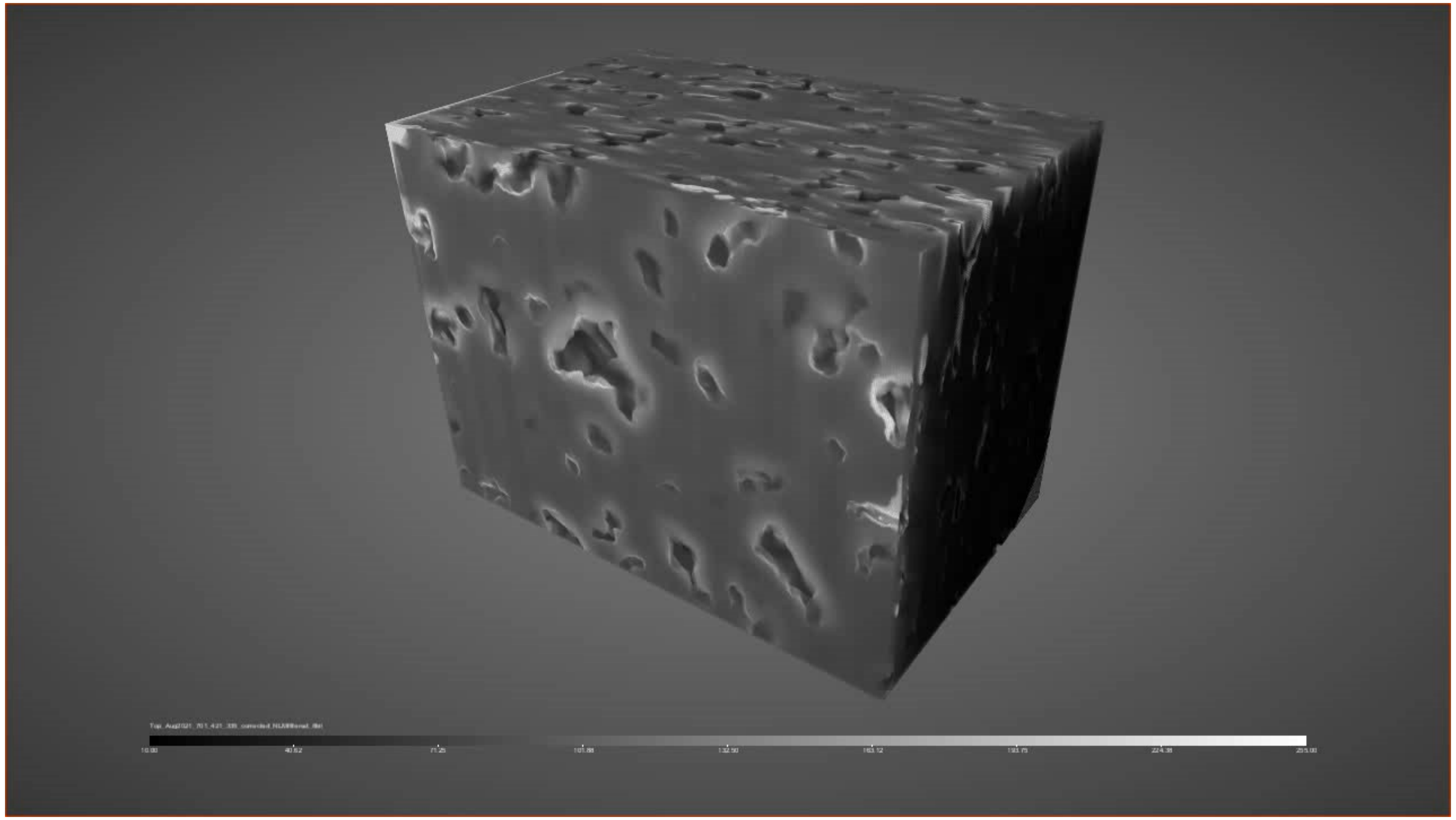
Part I: Porosity evolution studied by FIB-SEM tomography

Results: Example of 3D reconstruction



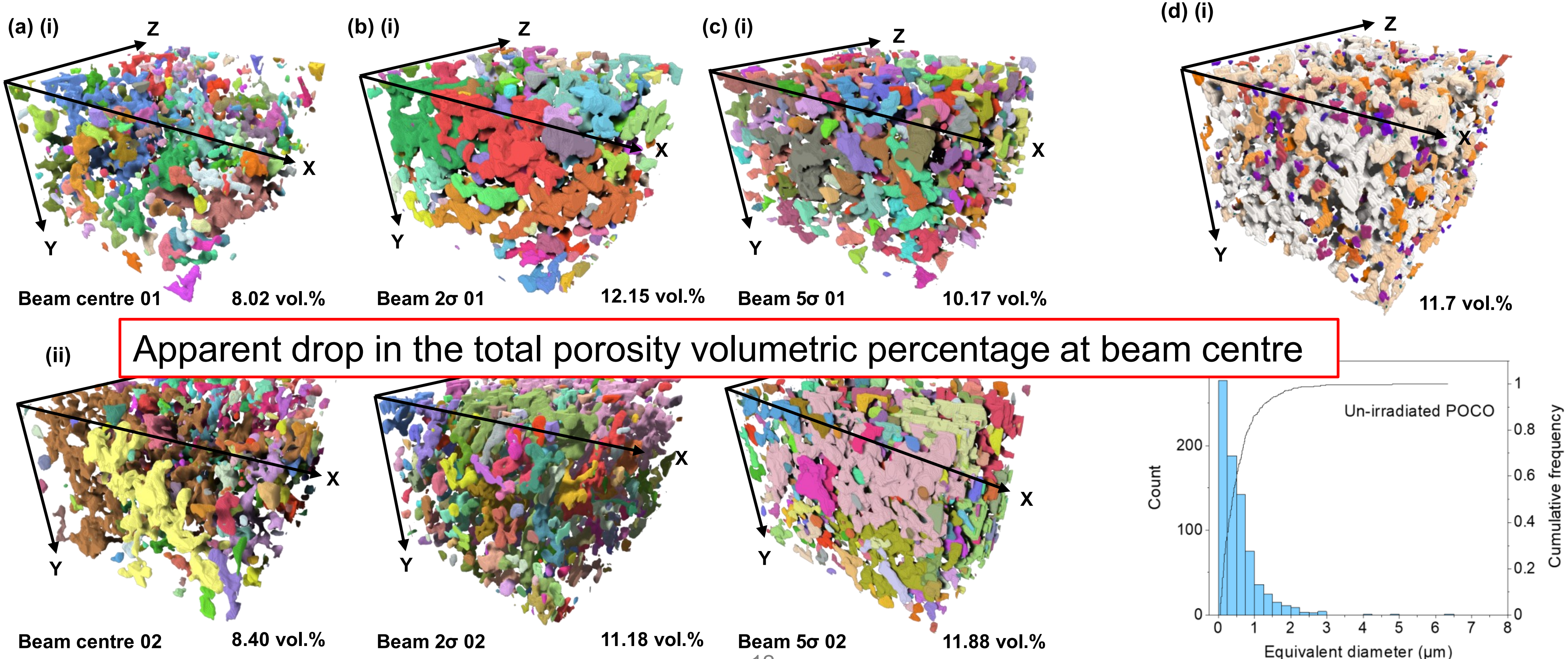
beam 5σ 02



beam 5σ 02

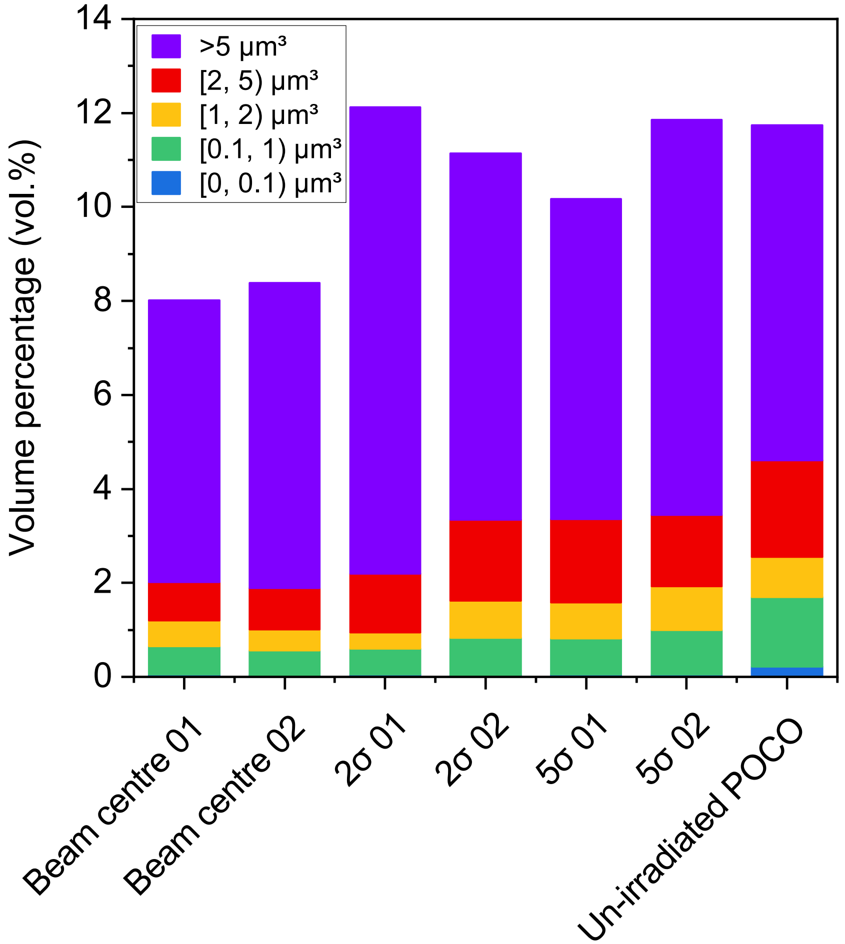
Part I: Porosity evolution studied by FIB-SEM tomography

Results: Porosity volume percentage



Part I: Porosity evolution studied by FIB-SEM tomography

Results: Porosity volume percentage



- Porosity volumetric percentage drop at beam centre is related to a volume reduction of pores having sizes greater than 0.1 µm^3
- It is these relatively larger pores that are heavily contributing the pore volumes within each rectangular space examined.
- The change is mostly likely to be caused by bulk dimensional swelling, but the underlying mechanism is still not clear and future work is needed.

Part I: Porosity evolution studied by FIB-SEM tomography

Conclusion:

- Total pore volume percentage decreased from ~ 12 vol.% to ~ 8 vol.% at proton beam centre area, primarily due to volume reduction of pores having sizes greater than 0.1 μm^3 due to dimensional swelling.
- A series of discussions have been made in our manuscript. But the underlying mechanism causing this reduction is still not clear due to such complicated irradiation environment. Future work on nanoscale structural change by TEM and diffraction techniques is planned.
- We are in the middle of revising our manuscript now addressing the reviewers' comments.
- However, a novel method using Raman spectroscopy seems to provide some insight into the crystal damage levels.

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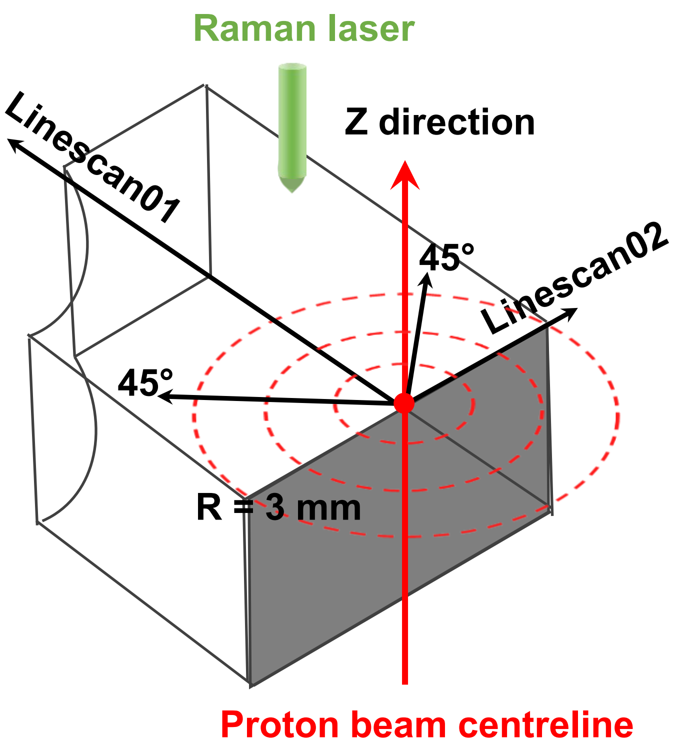
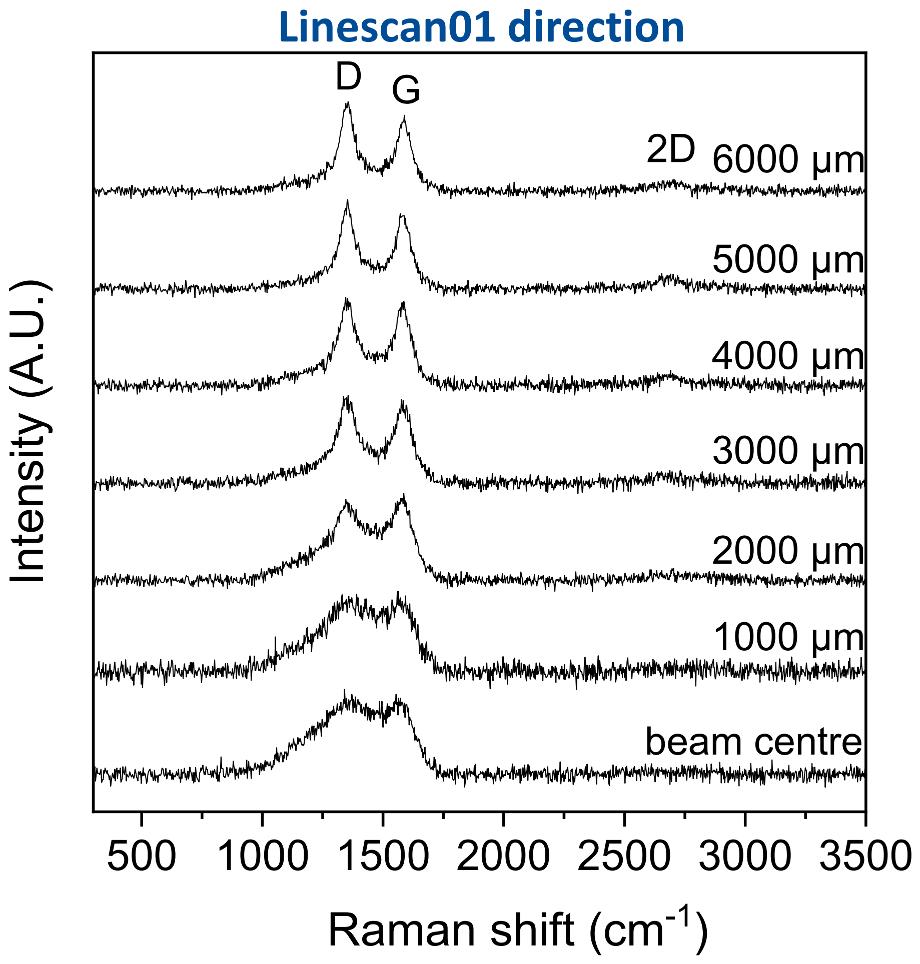
- **Part II: Crystal damage studied by Raman spectroscopy**
  - **Results**
  - **Conclusion**
- Future work

Part II: Crystal damage studied by Raman spectroscopy

Results: Spectral evolution



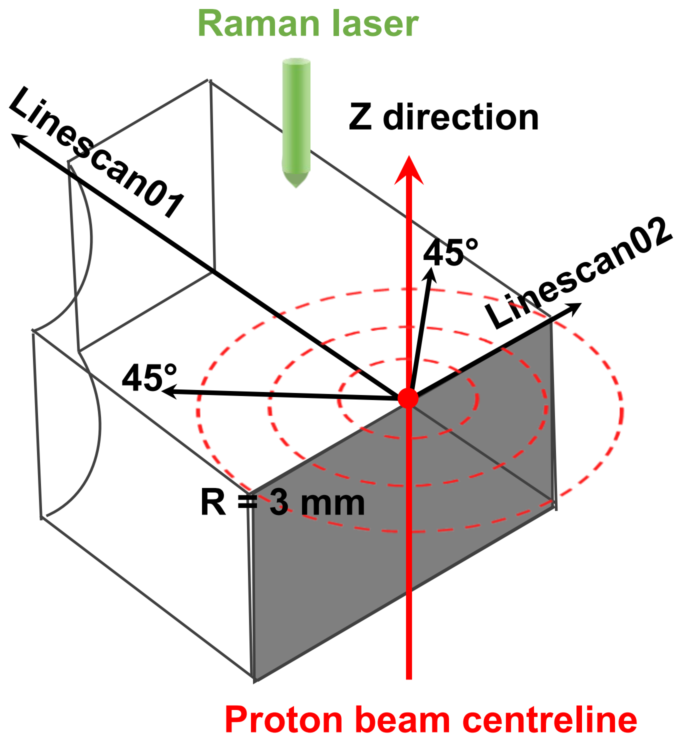
Schematic of the sample



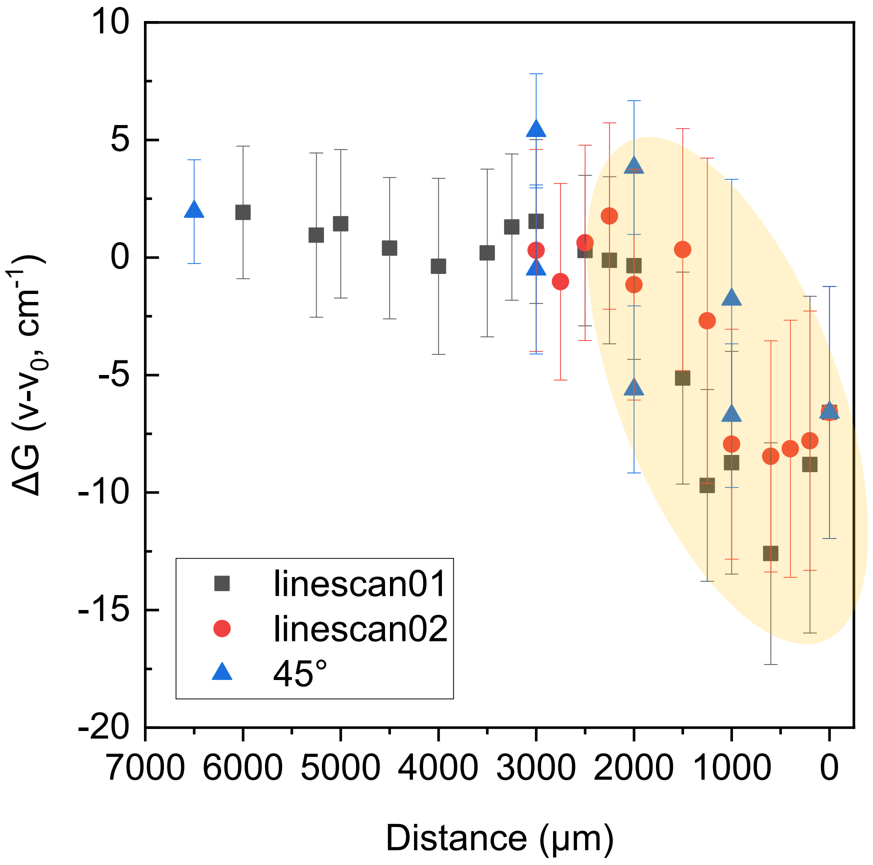
- D- and G-band broaden closer to beam centre.
- Second order 2D-band gradually disappear.
- Similarly for linescan02 and 45° directions.

Part II: Crystal damage studied by Raman spectroscopy

Results: Relative G-band shift (ΔG)



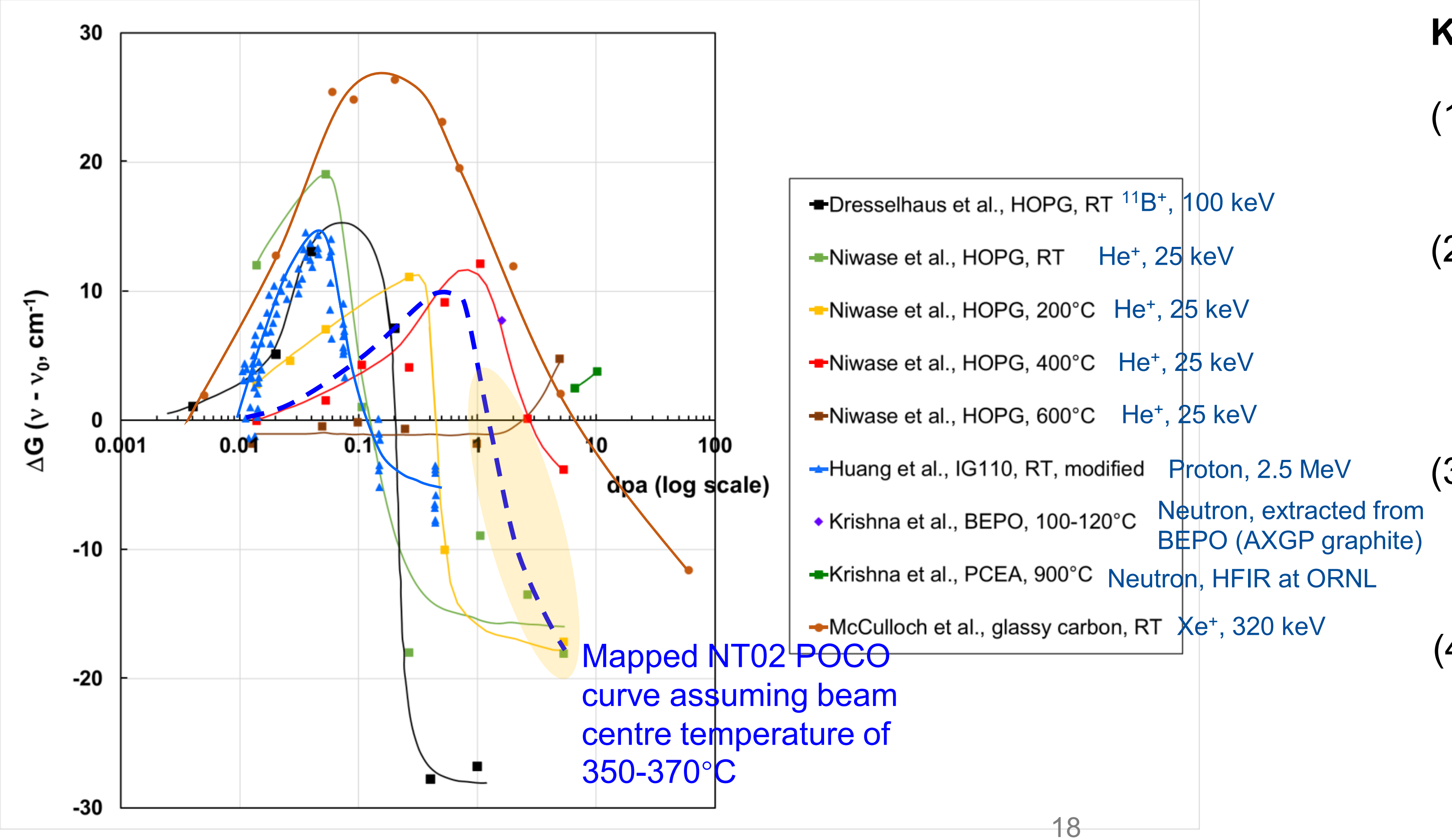
Schematic of the sample



- ΔG is the difference between measured G-band positions from this sample and that from non-irradiated POCO (~1583 cm^{-1}).
- ΔG mainly between +10 cm^{-1} and -20 cm^{-1} with a downward shifting trend within 3σ radius (3300 μm).
- ΔG within 2σ radius mainly between ~ 5 cm^{-1} and -20 cm^{-1}.

Part II: Crystal damage studied by Raman spectroscopy

Results: Relative G-band shift (ΔG)



Key messages:

(1) Consistent G-band position shift behaviour for all.

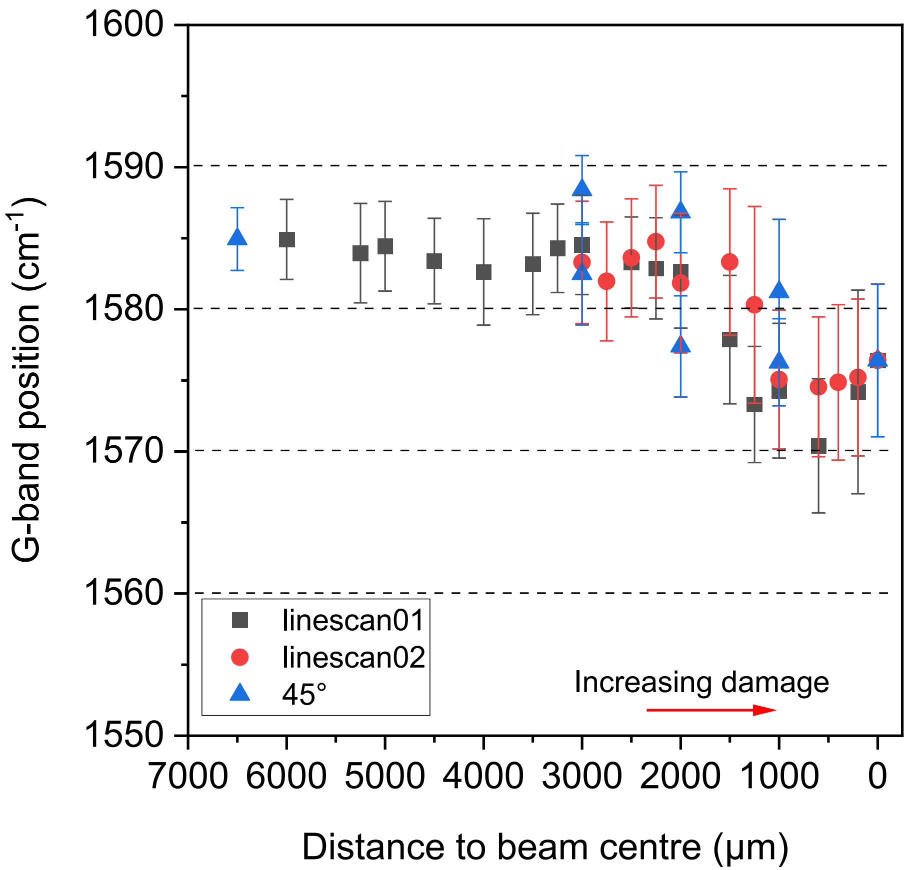
(2) G-band always shifts to higher frequencies then it saturates and shifts back downwards, i.e., a ‘bell’-shaped curve with a ‘turn-around’ peak.

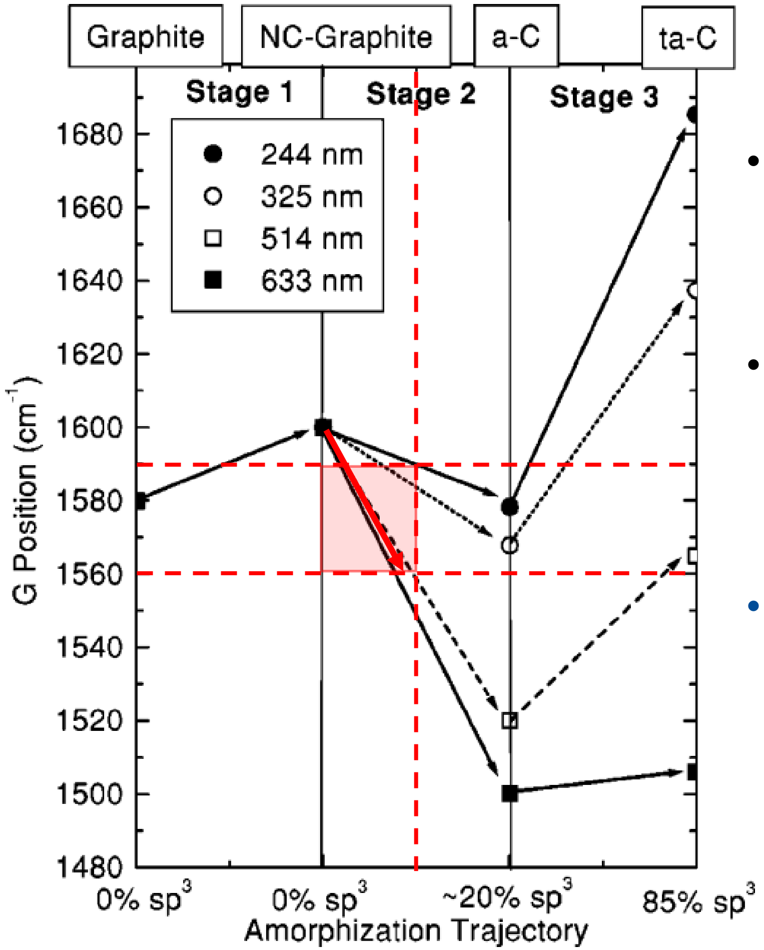
(3) The ‘turn-around’ peak delays towards higher dpa values with higher temperature.

(4) Estimated total damage level at beam centre 2σ is equivalent to ~2-5 dpa assuming peak temperature is ~ 350 – 370 °C.

Part II: Crystal damage studied by Raman spectroscopy

Results: G-band position shift





- Our data mainly within 1590 cm^{-1} to 1560 cm^{-1}.
- Correlating to Ferrari's three-stage amorphisation trajectory model.
- Approaching the middle point of stage 2, transitioning from nanocrystalline graphite (NC-Graphite) into amorphous carbon (a-C).

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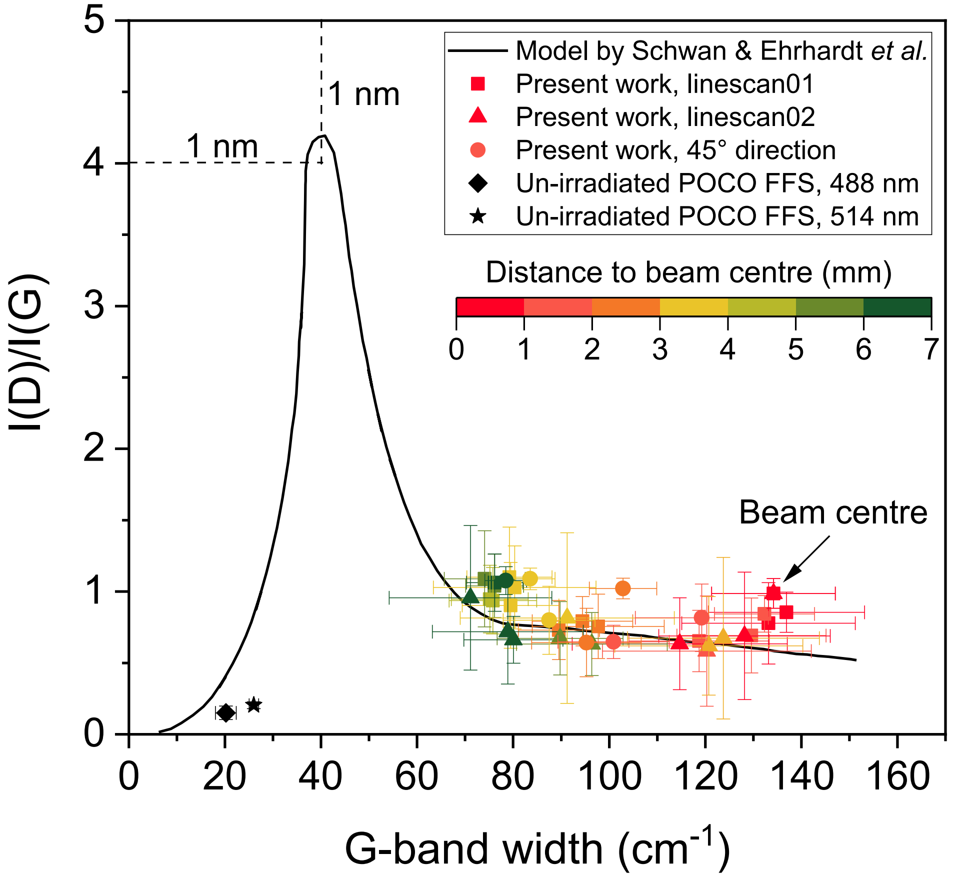
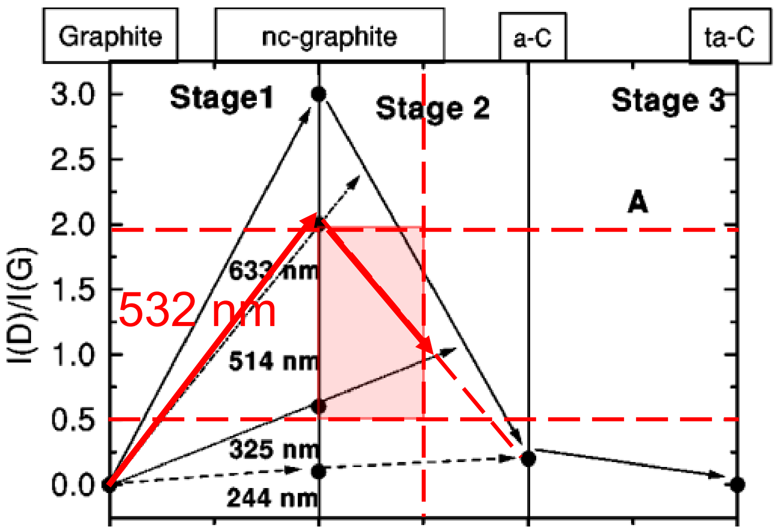
Part II: Crystal damage studied by Raman spectroscopy

Results: *I(D)/I(G)* ratio against G-band width

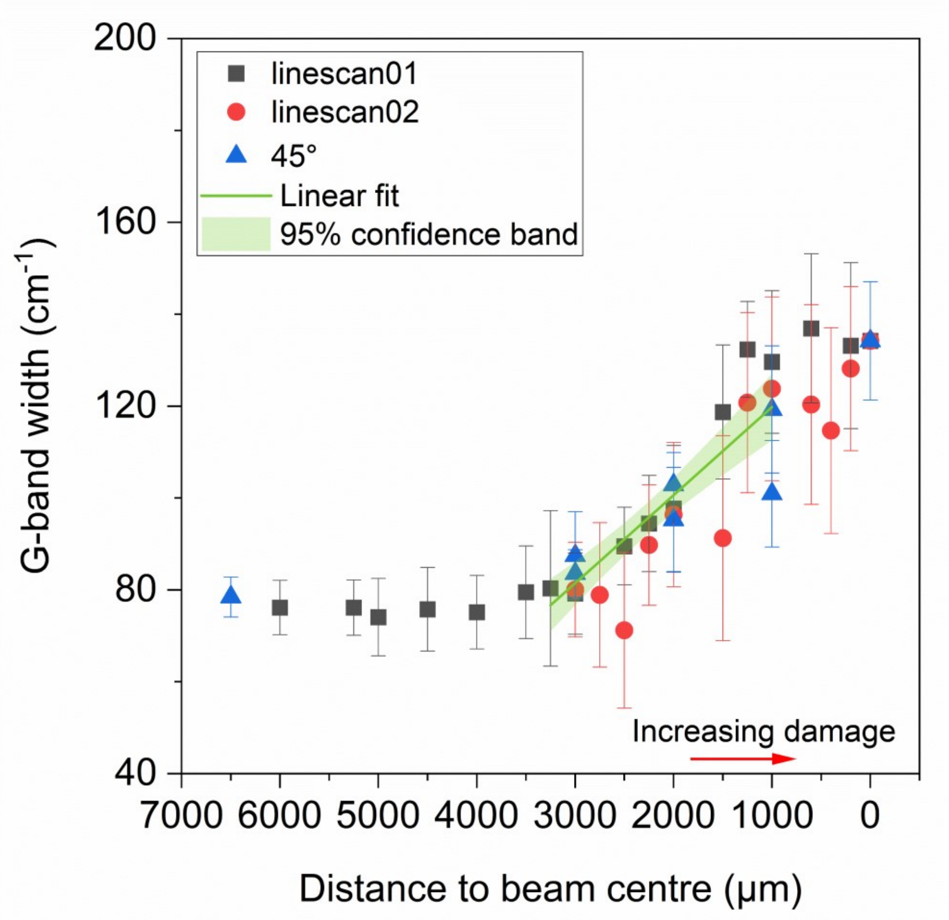




- *I(D)/I(G)* ratio does not further increase with increasing damage now.
- Our *I(D)/I(G)* ratios are mainly within 0.5 - 2.
- Instead, higher level of irradiation damage at beam centre is accompanied by more significant broadening (FWHM).
- Confirming again it is approaching the midway of stage 2, transitioning from NC-Graphite into a-C.

Part II: Crystal damage studied by Raman spectroscopy

Results: G-band width (FWHM)



(1) Linear broadening of width (W) within 1σ and 3σ distance (D).

(2) $W = -0.019D + 139$ (1000 μm≪D≪3300 μm)

(3) Broadening at a magnitude of 0.019 cm^{-1}/μm.

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Part II: Crystal damage studied by Raman spectroscopy

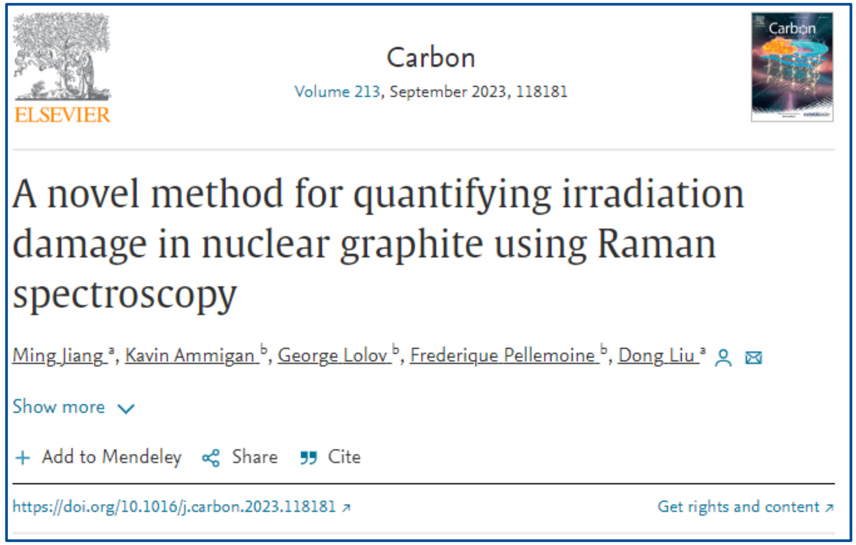
Conclusion:

- Damage level within proton beam 2 σ radius has been quantified to be ~ 2 - 5 dpa at ~ 350 – 370 °C. Correlating our NT02 POCO data to literature and the amorphization trajectory model gives a clear picture of the disorder stage and how much crystal damage it can further accommodate.

- It is possible to use the developed correlation technique as a lifetime assessment tool for these target graphites, alongside with regular non-destructive inspections in future.

- This method could potentially 'unify' irradiation damages across different grades of graphite subjected to different irradiation conditions, without the need for decoupling individual contributions.

- This work has just been published in the journal of *Carbon*.



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Volume 213, September 2023, 118181

A novel method for quantifying irradiation damage in nuclear graphite using Raman spectroscopy

Ming Jiang [a], Kavin Ammigan [b], George Lolov [b], Frederique Pellemoine [b], Dong Liu [a]

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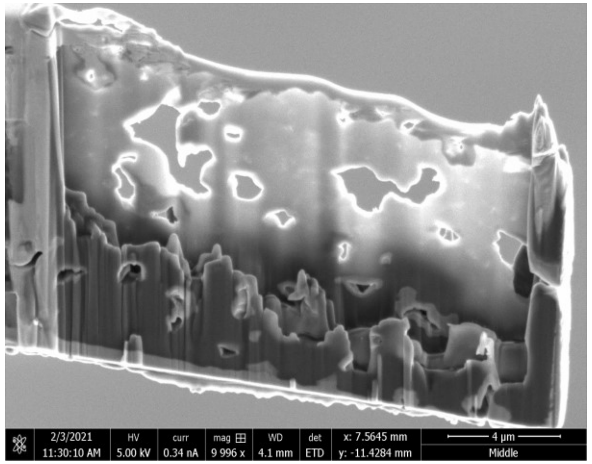
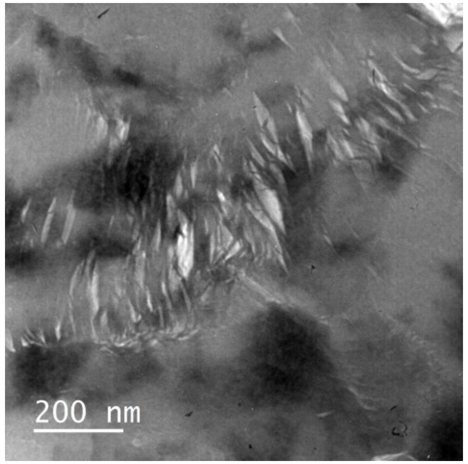
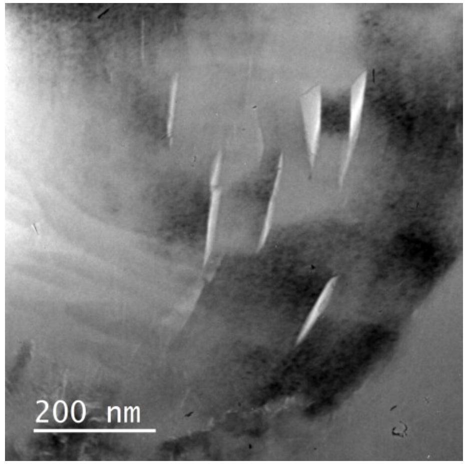
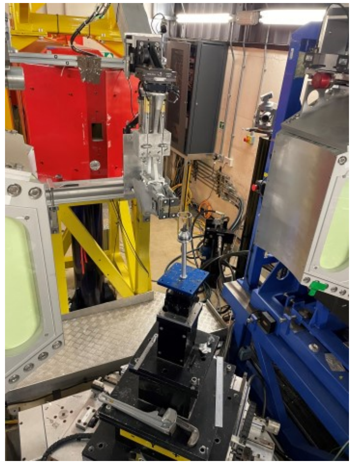
Future work

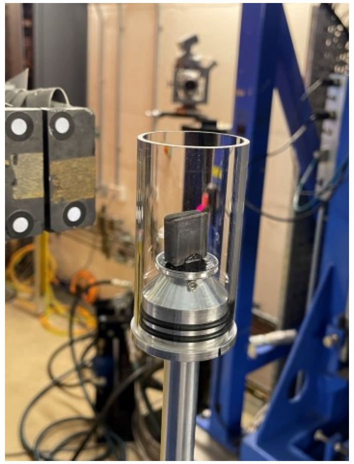
- Detailed TEM analysis of the thin foils extracted from exactly the same locations (ongoing).
- Beamline neutron diffraction for strain mapping in upstream POCO fins (data analysis next).
- My post-doc work with Science and Technology Facilities Council (STFC) and possibly Fermilab:
  - Help identify other high-density fine-grained graphite (assist DUNE and T2K).
  - Other grades (compared with IG430 and POCO) with proton irradiation experiment at Birmingham Cyclotron or Dalton Cumbrian Facility (UK).
  - PIE can be carried out at UKAEA, STFC and Bristol.











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