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Correlating Automated High-Throughput ADF-STEM and 4D-STEM Imaging for the Characterization of Irradiation-Induced Defects

Alex Lin[1], Sean H. Mills[1,2], Alexander Pattison[1], Wolfgang Theis[3], Andrew Minor[1,2] and Peter Ercius[1*]

[1.] National Center for Electron Microscopy, Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA, United States.
[2.] Department of Materials Science and Engineering, University of California, Berkeley, CA, United States.
[3.] Nanoscale Physics Research Laboratory, School of Physics and Astronomy, University Birmingham, Birmingham, United Kingdom.
* Corresponding author: percius@lbl.gov

Structural materials used in nuclear reactor environments are exposed to coupled extremes such as irradiation, high temperature, and corrosion which act synergistically to degrade their performance [1,2]. These degradation processes are directly limited or accelerated by irradiation-induced defects such as vacancies, interstitials, voids, and gas-filled bubbles [3]. However, it is challenging to decipher how different kinds of defects develop and interact with each other to lead to the failure of materials, as a mechanistic understanding of the associated thermodynamics and kinetics remains largely unknown.

A close inspection and tracking of point defects such as vacancies and interstitials is essential for understanding material behavior in the coupled irradiation and corrosion environment. The correlation of scanning electron nanobeam diffraction (or 4D-STEM [4]) and atomic resolution annular dark field scanning transmission electron microscopy (ADF-STEM) imaging has provided an opportunity for mapping defect distributions at the nanoscale and their associated strains, both of which have far-reaching implications for detailed analysis of complex irradiation and corrosion damage. Furthermore, automated high-throughput ADF-STEM imaging has been implemented to acquire atomic resolution ADF-STEM images autonomously over large fields of view (100 – 500 nm). This enables direct correlation of atomic resolution ADF-STEM images with the large field of view 4D-STEM strain maps, enabling a more statistically robust characterization of distributions and strain associated with a large number of individual defects.

He-ion beam irradiation, which has been extensively used to evaluate the tolerance of materials to extreme radiation environments, was performed on local regions in a [001] oriented Au thin film. Here, 4D-STEM with an ultrafast 4D Camera [5] was implemented to study the He-irradiated Au film and the strains accompanying nanoscale irradiation-induced defects were mapped using strain mapping methodology. Using a bullseye condenser aperture [6], this approach precisely measures local changes in the lattice *in situ* to study regions of the samples exposed to low He-ion irradiation dose (100 – 1000 ions/nm^2). The 4D-STEM strain maps reveal a high density of 1 – 5 nm defects in the irradiated zones (at a minimum probe size of 1 nm) over a field of view of 100 – 500 nm. By quantifying the strain and defect density in both damaged and pristine regions, the relationship between irradiation-induced defects and pre-existing defects such as stacking faults and twin boundaries can be further clarified.

Full quantification of strain associated with the irradiation-induced defects approaches the limit of 4D-STEM nanobeam imaging capabilities (~ 1 nm spatial resolution), so complimentary automated high-throughput ADF-STEM imaging was conducted on the regions previously imaged with 4D-STEM. In addition to automated microscope stage movements and microscope defocus adjustments, the high-throughput imaging workflow also acquired ADF-STEM image 0-90° pairs that are subsequently used for 2D Gaussian peak fitting to correct scan distortions and precisely locate the positions of atomic columns [7]. This allows for the quantitative characterization of atomic displacements and changes in the lattice parameter in and around defect clusters within the irradiation damaged zones. This correlative approach to characterizing point defects through maximizing resolution (HR-STEM) and expanding field of view (4D-STEM) fundamentally improves the understanding of complex irradiation damage mechanisms and provides a new pathway for engineering materials in future nuclear energy systems [8].

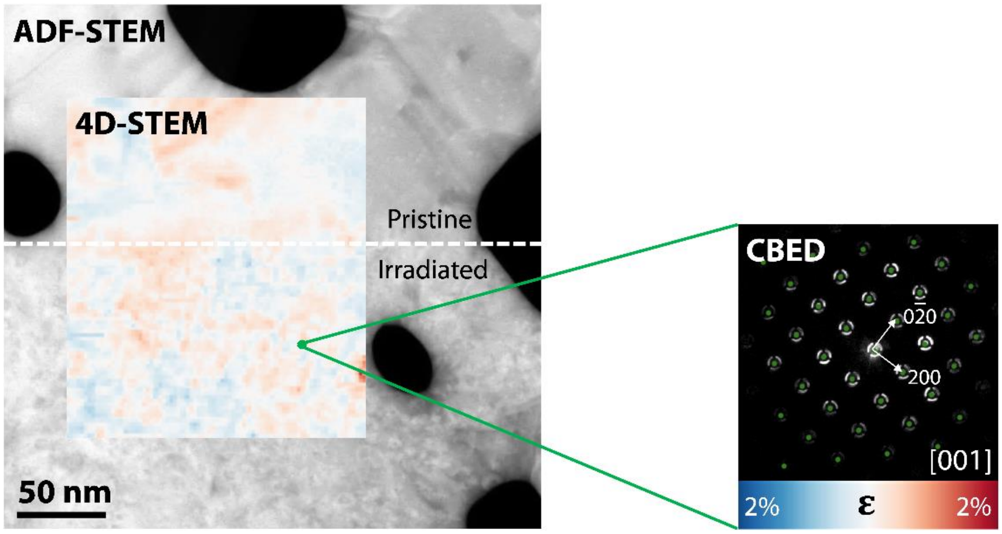

Figure 1. 4D-STEM characterization using a bullseye condenser aperture. The strains accompanying nanoscale irradiation-induced defects were mapped using diffraction disks in the convergent beam electron diffraction (CBED) patterns.

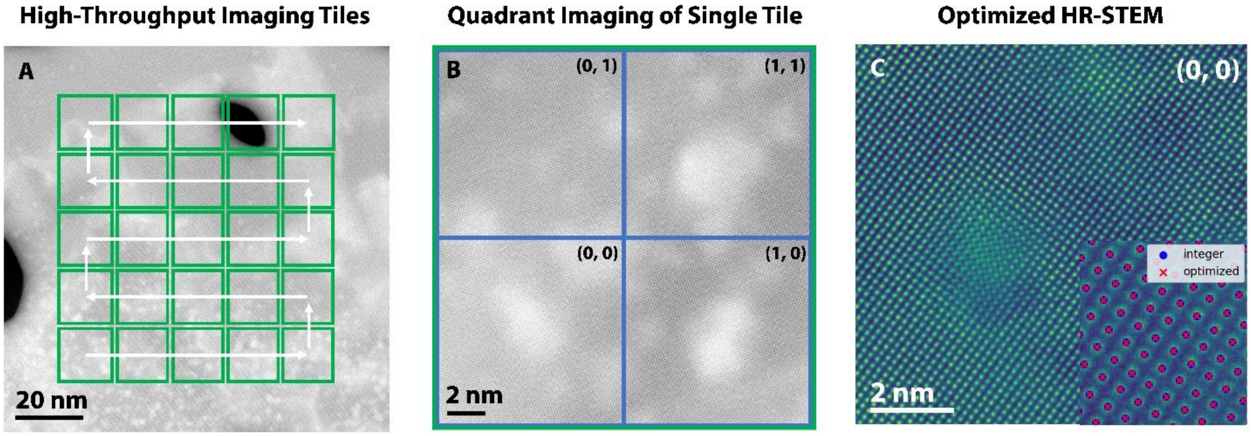

Figure 2. Automated high-throughput ADF-STEM imaging workflow. (A) Imaging tiles are automatically created based on image magnification. Arrows show the directions of the microscope stage movements. (B) Each imaging tile is split into a quadrant for the acquisition of high-resolution ADF-STEM images in each square. (C) 2D Gaussian peak fitting optimizes the HR-STEM images for locating precise locations of atomic columns.

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