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In situ 4D-STEM Imaging to Develop a Fundamental Understanding of Coupled Transport of Vacancies

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Extreme nuclear reactor environments require materials to maintain their integrity all while a range of processes act in unison to degrade their performance. These processes are directly limited or accelerated by defects produced under irradiation. Until recently, capabilities to directly measure concentration of vacancies in metals have been limited to bulk techniques such as x-ray diffraction [1] or positron annihilation spectroscopy [2]. There is still a lack of established techniques that enable direct mapping of point defects using electron microscopy. However, recent developments in four-dimensional scanning transmission electron microscopy (4D-STEM) with high-speed direct electron detectors [3, 4] and high resolution STEM provide an opportunity for potentially mapping vacancy distributions at the nanoscale and their associated strains, both of which have far-reaching implications for detailed analysis of complex damage and accumulation of point defects.

The present experiment utilizes a Au thin film as a model to demonstrate a method for measuring vacancy concentration via 4D-STEM by closely following the differential thermal expansion method of measuring concentrations of point defects [1]. Figure 1 presents measurement of lattice parameters and defect distributions conducted *in situ* as a function of increasing temperature on suspended Au thin films. The concentration of thermally generated (equilibrium) vacancy defects reaches a peak (~10^{-4}) in the high temperature regime (17ºC – 1000ºC in Au) [4, 5] causing a nonlinear separation between measured volume expansion (previously via optical dilatometry, but now HR-STEM) and lattice expansion (previously via XRD, but now 4D-STEM). The vacancy concentration at room temperature varied after multiple short-term heating cycles suggesting incomplete relaxation in the thin film which is attributed to limited defect mobility. Moreover, annealing of defects over a long-term hold at room temperature was observed. The vacancy mapping methodology was then applied to non-equilibrium defects accumulated in pure Al via knock-on electron beam irradiation. Furthermore, this technique is relatively insensitive to subtle changes in orientation and can be collected over a large (100 – 500 nm) field of view. This combined approach to characterizing point defects through maximizing resolution and field of view (HR-STEM and 4D-STEM) fundamentally improves the understanding of nanoscale defect accumulation under extreme thermal and irradiated conditions and provides a new pathway for engineering materials in future nuclear energy systems [6].

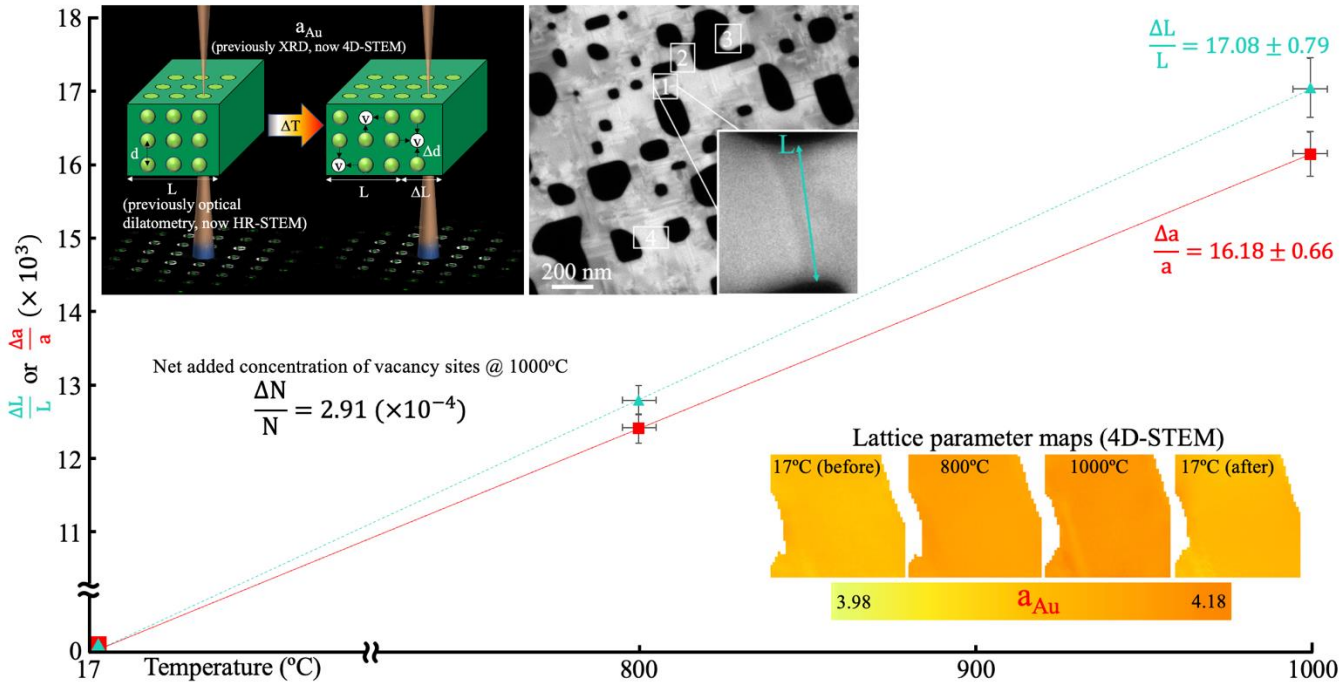

Figure 1. Length expansion (cyan triangles, dashed cyan line) and lattice parameter expansion (red squares, solid red line) of pure Au thin film with respect to temperature. (inset, top left) Schematic illustration of nano-beam electron beam converged on the Au thin film. A pure crystal at 17ºC which is free of defects with a geometric dimension L and initial d-spacing (reference lattice). To the right is a crystal at high temperature containing vacancy defects (white positions with a "v"). The change in lattice parameter and d-spacing are measured via HR-STEM and 4D-STEM, respectively. (inset, top center) Length measurements are conducted on HAADF-STEM images of most narrow point of ligament sections (white boxes) corresponding to the 4D-STEM measurement. (inset, bottom right) Lattice parameter maps derived from 4D-STEM measurements at varied temperature for a single heat treatment cycle. Scale bar of lattice parameter maps shown in Angstroms.

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