Temperature Dependence of Helium-Implantation-Induced Lattice Swelling in Polycrystalline Tungsten: X-ray Micro-Diffraction and Eigenstrain Modelling

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Using synchrotron X-ray micro-diffraction and Eigenstrain analysis the distribution of lattice swelling near grain boundaries in helium-implanted polycrystalline tungsten is quantified. Samples heat-treated at up to 1473 K after implantation show less uniform lattice swelling that varies significantly from grain to grain compared to as-implanted samples. An increase of lattice swelling is found in the vicinity of some grain boundaries, even at depths beyond the implanted layer. These findings are discussed in terms of the evolution of helium-ion-implantation-induced defects.

Keywords: helium ion-implantation; X-ray micro-diffraction; tungsten; lattice swelling; Eigenstrain analysis

Tungsten alloys are likely candidates for plasma facing components in future nuclear fusion reactors. They will be exposed to high temperatures (>1500 K), high neutron flux (~ 2 MW/m²) and intense hydrogen and helium bombardment (~ 10 MW/m²) [1]. Mechanical properties are strongly affected by the interaction of retained helium with irradiation-induced defects, which can be studied efficiently using helium ion implantation [2]. In metals defect microstructure is dominated by the propensity of helium to form tightly bound configurations with vacancies [3, 4]. Below ~ 550 K vacancies in tungsten are immobile and do not cluster [5]. Indeed previous transmission electron microscopy (TEM) of tungsten implanted with 3000 atomic parts per million (appm) of helium at 573 K showed no visible defects [2], suggesting that all vacancy and self interstitial atom (SIA) defects were below the TEM resolution (nominally 0.25 nm).

Above ~550 K vacancies become mobile, and clustering of vacancies and SIAs occurs [6]. These defects are also expected to migrate deeper into the material, beyond the implanted layer, escape at free surfaces and accumulate at grain boundaries. Helium accumulation at grain boundaries, leading to embrittlement, is a particular concern [7].

We explore experimentally the migration of defects in helium-implanted tungsten at fusion relevant temperatures. Synchrotron X-ray micro-beam Laue diffraction (Fig.1(a)) allows the probing of helium-implantation-induced defects, invisible in TEM, by measuring the associated lattice distortions [8]. We use this technique to quantify near grain boundary lattice strains in polycrystalline tungsten implanted with helium at 298 K, followed by heat treatment at up to 1473 K. Experimental data is interpreted using an Eigenstrain modelling approach to recover lattice swelling and allow conclusions about defect migration to be reached.

Three tungsten slices (99.99%), recrystallized in vacuum at 1673 K, were mechanically polished to a flat, defect-free finish. Electron Backscatter Diffraction (EBSD) showed large grains (30 – 300 μm) with negligible texture. Helium ion implantation at 298 K (National Ion Beam Facility, UK) used 13 ion energies from 0.05 – 2.0 MeV (Supplementary Material) to achieve a near uniform calculated helium concentration of 3180 ± 220 appm within a 2.7 μm thick surface layer. The implantation profile, predicted using the Stopping Range of Ions in Matter (SRIM) code [9] (68 eV displacement threshold [10]) is shown in Fig.1(b). The calculated displacement damage at depths <2.7 μm is 0.21 ± 0.03 displacements per atom (dpa). Sample T298 remained as implanted. Samples T1273 and T1473 were heat treated for 12 hours in vacuum at 1273 K and 1473 K respectively.

Micro-beam Laue measurements used beamline 34-ID-E at the Advanced Photon Source (APS), Argonne National Lab, USA. Fig.1(a) shows a schematic of the experimental setup [11, 12]. The polychromatic X-ray beam (7 – 30 keV) was focussed to $0.4 \times 0.6 \mu\text{m}^2$ (horizontal x vertical full width at half maximum (FWHM)) on the sample. Laue diffraction patterns, containing 20+ peaks per grain, were analysed using LaueGo (tischler@anl.gov) to determine lattice orientation and deviatoric elastic strain tensor, e^* , defined as $e^* = e_T - (1/3)\text{tr}(e_T)\mathbf{I}$, where e_T is the total lattice strain and $\mathbf{I}$ the identity matrix. Differential Aperture X-ray Microscopy (DAXM) with a 50 μm diameter platinum wire allowed a nominal spatial resolution of 0.5 μm along the incident beam direction [13, 14].

Fig.1(c) shows depth-resolved deviatoric strains measured in T298 (mean and standard deviation computed from measurement in Fig.2(b)). Beyond the implanted layer all strain components are small. In the implanted layer (depths < 2.7 μm), out-of plane deviatoric strain is large and positive ($e_{zz}^* = (7.6 \pm 1.3) \times 10^{-4}$), whilst the in-plane deviatoric strains are similar in magnitude and negative ($e_{xx}^* = (-3.7 \pm 1.3) \times 10^{-4}$; $e_{yy}^* = (-3.9 \pm 1.9) \times 10^{-4}$). This suggests swelling of the implanted layer. Compatibility of the unimplanted substrate and the implanted layer requires in-plane total lattice strains to be zero ($e_{T,xx} = e_{T,yy} = 0$). As tungsten is elastically isotropic at room temperature [15], $e_{T,zz} \approx e_{zz}^* - (e_{xx}^* + e_{yy}^*)/2$. The total lattice strain, e_T , is the superposition of elastic strain, e , and volumetric strain due to defects, e_v , such that $e_T = e + (1/3)e_v \mathbf{I}$. Given the present boundary conditions, lattice swelling, e_v , is:

$$e_v = \frac{3(1-\nu)}{1+\nu} e_{T,zz}, \quad (1)$$

where the Poisson ratio $\nu = 0.28$ [15]. For T298 $e_v = (1.93 \pm 0.49) \times 10^{-3}$, which is in good agreement with measurements of a helium-implanted tungsten-rhenium alloy [8]. The prefactor in eqn.1 arises due to the boundary constraints in our sample, as e_v refers to the lattice swelling due to defects that would be observed in an unconstrained sample [8].

To probe lattice swelling near grain boundaries, two boundaries per sample were selected (straight, length > 100 μm , near-perpendicular to sample surface). At each boundary DAXM measurements were carried out along a line perpendicular to the boundary (Fig.2(a)), with the measurement plane inclined at 45° to the sample surface (Fig.1(a)). Since the grain boundaries are long, strains could be projected onto a vertical slice through each grain boundary, with 20 μm width parallel to the sample surface (41 measurement points) and 9 μm height in the depth direction (27 measurement points).

In T298 an increase in e_{zz}^* near the surface due to helium-implantation-induced lattice swelling is evident (Fig.2(b)). The profile varies little with distance from the grain boundary. The minor axis width of Laue diffraction peaks captures lattice distortion spread within the gauge volume [16]. In T298 (Fig.2(c)) peak width is substantially increased in the implanted

layer due to implantation-induced point defects. The extent of this region agrees well with the lattice strain profile (Fig.2(b)) and the SRIM-predicted implantation profile (Fig.1(b)).

Determining the lattice swelling distribution that causes a measured deviatoric strain distribution becomes less straightforward when lattice swelling is not only a function of depth (T298), but also of position perpendicular to the grain boundary (T1273 and T1473). We develop an Eigenstrain approach [17] to solve the inverse problem of recovering the lattice swelling distribution from a measured deviatoric strain distribution. Eigenstrain refers to a permanent, heterogeneous inelastic strain distribution (here defect-induced lattice swelling) that causes a non-uniform residual elastic strain field. Total strain is the sum of Eigenstrain, ϵ^* , and residual elastic strain, e . We measured residual deviatoric elastic strain, e^* , at N points x_i . $\epsilon^*(x)$ denotes the unknown Eigenstrain distribution at position x that creates an unknown deviatoric elastic strain field $E^* = f(\epsilon^*)$. We seek an Eigenstrain distribution, $\epsilon^*(x)$, such that:

$$J = \sum_{i=1}^N [\mathbf{e}_{kl}^*(x_i) - \mathbf{E}_{kl}^*(x_i)]^2 \quad (2)$$

is minimised. This summation can be extended to include several strain components kl . The unknown Eigenstrain distribution is approximated as:

$$\epsilon^*(x) = \sum_{j=1}^M c_j \xi_j(x), \quad (3)$$

where $\xi_{j=1..M}$ are M basis functions and $c_{j=1..M}$ are unknown coefficients. Each basis function produces a residual deviatoric elastic strain field $\mathbf{E}_j^*(x)$. Using superposition we obtain

$$\mathbf{E}_{kl}^*(x) = \sum_{j=1}^M c_j \mathbf{E}_{kl,j}^*(x). \text{ To solve this inverse Eigenstrain problem requires a set of}$$

coefficients $c_{j=1..M}$ that minimise eqn. 2.

The chosen Eigenstrain basis functions correspond to a local swelling of 10^{-6} imposed at each measurement point, within a volume equal to the measurement gauge volume ($\xi_j(x) = 1/3 \times 10^{-6} \mathbf{I}$; $\mathbf{I}$ is the identity matrix). The determined coefficients $c_{j=1..M}$ correspond to volumetric lattice strain, e_v , in microstrain (10^{-6}).

Deviatoric elastic strain fields, $\mathbf{E}_j^*(x)$, were computed using CAST3M (<http://www-cast3m.cea.fr>) with QUA4 elements. Far from the grain boundary we imposed $e_{T,xx} = 0$ to enforce compatibility between implanted layer and unimplanted substrate. For long straight grain boundaries variation along the grain boundary is negligible ($e_{T,yy} = 0$). $e_{T,zz} = 0$ was imposed at 20 μm depth, much deeper than the implanted layer. Results were insensitive to exact boundary conditions provided they were imposed sufficiently far from the intersection of grain boundary and sample surface.

Several fitting schemes were considered (Supplementary Material). Below, e_{xx}^* and e_{zz}^* were taken into account when determining coefficients c_j . Tikhonov regularization [18] was used to reduce the influence of experimental noise by penalising the gradient of lattice swelling in both x and z directions. Thus we minimised a more complicated expression for J than eqn. 2:

$$J = \|\mathbf{Ac} - \mathbf{b}\|_2^2 + \lambda \left(\|\nabla_x c\|_2^2 + \|\nabla_z c\|_2^2 \right), \quad (4)$$

$$\text{where } \mathbf{A} = \begin{bmatrix} (E_{xx,j}^*(x_i))_{i=1..N, j=1..N} \\ (E_{zz,j}^*(x_i))_{i=1..N, j=1..N} \end{bmatrix} \text{ and } \mathbf{b} = \begin{bmatrix} (e_{xx}^*(x_i))_{i=1..N} \\ (e_{zz}^*(x_i))_{i=1..N} \end{bmatrix}. \nabla_x c, \nabla_z c \text{ are the}$$

gradients of the scalar field of coefficients c_j in the x and z directions respectively. Since lattice swelling implies an expansion of the lattice we require $c_j \geq 0$. This was enforced using the *lsqnonneg* function in Matlab to minimise eqn. 4. In eqn. 4, $\lambda = 2.6 \times 10^{-7}$, as determined by considering experimental uncertainties (Supplementary Material).

The reconstructed e_{zz}^* strain field from Eigenstrain analysis of boundary T298_B1 and the corresponding Eigenstrain distribution are shown in Fig.2(d),(e) respectively. Agreement between the experimentally measured (Fig.2(b)) and reconstructed (Fig.2(d)) e_{zz}^* strain fields is excellent. Tikhonov regularisation makes the reconstructed strain field a low-pass-filtered version of the measured one, whilst retaining the salient features. The Eigenstrain map (Fig.2(e)) shows localised swelling in the implanted layer. The average value of e_v up to 2.7 μm depth is $(1890 \pm 340) \times 10^{-6}$, in agreement with our previous estimate of $(1930 \pm 490) \times 10^{-6}$ from Fig.1(c).

Fig.3 shows Eigenstrain maps reconstructed for two measurements each in T298 (Fig.3(a),(b)), T1273 (Fig.3(c),(d)) and T1473 (Fig.3(e),(f)). All maps show volumetric lattice strain, $e_v = \varepsilon^*(x)$, on the same scale range.

Swelling of the implanted layer in T298 is homogeneous and largely confined to the SRIM-predicted implantation depth of $\sim 3 \mu\text{m}$. In comparison both maps in T1273 (Fig.3(c),(d)) show a reduction of swelling and an increase in heterogeneity. One of the boundaries also shows an increase in near boundary swelling (Fig.3(c)), whilst no such localisation is seen in the other (Fig.3(d)). The T1473 grain boundaries (Fig.3(e),(f)) show a similar behaviour.

TEM [2], positron annihilation spectroscopy [3], thermal desorption spectroscopy [19] and ab-initio calculations [4] suggest that helium-filled vacancies and SIA defects will form under our implantation conditions (298K, He-ions with energies $\leq 2 \text{ MeV}$). At 298 K significant clustering of these defects is not expected. Even in tungsten implanted with helium at 573 K at similar ion energy and dose, TEM showed no visible defect clusters [2]. The uniform distribution of swelling in the implanted layer in T298 is consistent with damage dominated by uniformly distributed point defects much smaller than the sampling volume size ($\sim 0.5 \times 0.5 \times 0.5 \mu\text{m}^3$).

Both T1273 and T1473 show reduced, less uniform, lattice swelling than T298 (Fig.3). This can be understood as follows: Above $\sim 550 \text{ K}$ vacancies in tungsten become mobile [5] and clustering of SIAs and vacancies is expected. Lattice swelling is dominated by SIAs, whilst vacancies contribute little [8]. Swelling due to N isolated SIAs is $N\Omega_r^{\text{SIA}}$, where Ω_r^{SIA} is the SIA relaxation volume. For large N an SIA cluster will form a $1/2 \langle 111 \rangle$ prismatic dislocation loop with a volume approaching $N\Omega_0$, where Ω_0 is the atomic volume. As $\Omega_r^{\text{SIA}} > \Omega_0$ [8], SIA clustering reduces lattice swelling. Increased mobility of vacancies and SIAs at elevated temperatures also allows their migration to sinks (e.g. sample surface, grain boundaries), providing a second pathway for lattice swelling reduction. In T1473 (Fig.3(e),(f)) swelling also shows strong grain-to-grain variation, indicating that migration and clustering of helium-implantation-induced defects is highly dependent on lattice orientation.

Swelling at one grain boundary each in T1273 and T1473 (Fig.3(c),(e)) is significantly increased, suggesting the accumulation of defects during heat treatment. The other

boundaries in T1273 and T1473 (Fig.3(d),(f)) do not show this effect. No clear link could be established between grain boundary structure and increased swelling after heat treatment. However we can unambiguously state that at elevated temperatures irradiation-induced defects accumulate at some grain boundaries, even at depths beyond the implanted layer.

In summary our analysis revealed that helium-ion-implantation leads to significant swelling in tungsten. Samples implanted at room temperature showed uniform swelling confined to the implanted layer. After heat treatment above 1273 K lattice swelling became heterogeneous, indicating defect migration and clustering. Near some grain boundaries lattice swelling at depths beyond the implanted layer was observed, indicating that at elevated temperatures irradiation-induced defects can migrate deeper into the bulk. Further work will clarify whether this could compromise long-term structural integrity of plasma-facing components in future fusion reactors.

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Figure captions:

Figure 1: (a) Schematic of experimental configuration indicating positioning of X-ray beam, detector, scanning wire, sample, sample coordinates and alignment of the grain boundary. (b) Calculated helium implantation and displacement damage profiles. (c) Deviatoric direct strain profiles averaged over 41 measurement locations shown in Fig.2 (b), plotted as a function of depth.

Figure 2: T298 boundary B1: (a) Optical micrograph of boundary showing intersection of sample surface and measured slice (black line). (b) Measured out-of-plane deviatoric strain, e_{zz}^* . (c) Laue peak minor axis FWHM in pixels averaged over all peaks. (d) Out-of-plane deviatoric strain, e_{zz}^* , reconstructed by Eigenstrain. (e) Swelling, e_v , determined from Eigenstrain ($\times 10^{-6}$). Maps (b) – (e) are in the x-z plane and dashed lines mark the grain boundary.

Figure 3: Lattice swelling, e_v , determined from Eigenstrain ($\times 10^{-6}$). (a), (b) Sample T298. (c), (d) Sample T1273. (e), (f) Sample T1473. Maps are in the x-z plane and dashed lines mark grain boundaries.

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Figure 1

(a)

(b)

(c)

Figure 2

(a)

(b)

experimental e_{zz}^*

(c)

full width at half maximum

(d)

reconstructed e_{zz}^*

(e)

coefficients

Figure 3

Figure 3 displays six panels (a-f) showing AFM-IR intensity maps of a polymer surface. The panels are arranged in a 3x2 grid. The left column (a, c, e) shows the unheated sample, and the right column (b, d, f) shows the sample after heating. The rows correspond to different temperatures: (a, b) unheated, (c, d) $T = 1273\text{ K}$, and (e, f) $T = 1473\text{ K}$. Each panel includes a scale bar indicating $5\text{ }\mu\text{m}$ and a color scale ranging from 0 to 3000. A dashed line in each panel indicates a boundary or defect.