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Characterization of defect clusters in ion-irradiated tungsten by X-Ray diffuse scattering

Peihao Sun [a, b, *], Yongqiang Wang [c], Mungo Frost [a], Christopher Schönwälder [a, d], Abraham L. Levitan [a], Mianzhen Mo [a], Zhijiang Chen [a], Jerome B. Hastings [a], George R. Tynan [e, f], Siegfried H. Glenzer [a], Philip Heimann [g]

[a] SLAC National Accelerator Laboratory, Menlo Park, CA, 94025, USA
[b] Department of Physics, Stanford University, Stanford, CA, 94305, USA
[c] Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, NM, 87545, USA
[d] Friedrich-Alexander-Universität Erlangen-Nürnberg, 91054, Erlangen, Germany
[e] Center for Energy Research, San Diego, La Jolla, CA, 92093, USA
[f] Department of Mechanical and Aerospace Engineering, Jacobs School of Engineering, UC, San Diego, La Jolla, CA, 92093, USA
[g] Linac Coherent Light Source, SLAC National Accelerator Laboratory, Menlo Park, CA, 94025, USA

HIGHLIGHTS

- Size-distribution of defect clusters are obtained *via* X-ray diffuse scattering.
- More vacancies than interstitials are observed at each irradiation level.
- The diameter of vacancy loops increases with irradiation dose.

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ABSTRACT

Defect clusters in ion-irradiated tungsten are characterized by X-ray diffuse scattering. Single crystal tungsten samples are irradiated at 300 K by 0.5–5 MeV copper ions with total doses of 0.2, 0.6, 2, and 5 displacements per atom (DPA). Analysis of the diffuse scattering intensities allows the determination of the concentration of vacancy- and interstitial-type dislocation loops as a function of loop radius for each sample. More vacancies than interstitials are observed in each sample, and the radius of the vacancy loops increases with irradiation dose. For the 0.2DPA sample the average vacancy loop radius is 10.1±3.0 Å with a defect density (counting all atoms in the loops) of $(10.2\pm1.5)\times10^{-4}$ per tungsten atom, increasing to an average loop radius of 16.1±3.1 Å at a density of $(8.5\pm1.0)\times10^{-4}$ per tungsten atom for the 5DPA sample.

1. Introduction

In the design of next-step magnetic confinement fusion devices such as ITER and DEMO, tungsten is an attractive materials choice for the divertor and the plasma-facing wall [1]. As a result, it is subject to high levels of irradiation, both by large fluxes of hydrogen isotope ions with energies in the range of tens to hundreds of electronvolts as well as by energetic neutral atoms formed by charge exchange of thermal neutrals with plasma ions [2]. These particles implant in the near-surface region (tens to hundreds of nanometers deep), go into solution with the material, and then begin to diffuse. At the same time, in deuterium-tritium (D-T) burning plasma experiments, the tungsten material will also be subjected to a significant flux of 14 MeV neutrons. Collisions between these energetic neutrons and lattice atoms then induce displacement cascades that relax into multiple vacancy-interstitial pairs. These pairs subsequently evolve into larger defects such as dislocation loops and voids. In multicrystalline materials, these damage-induced defects are created within a background of pre-existing, or intrinsic, defects associated with grain boundaries, crystal defects, and impurities. These intrinsic and damage-induced

* Corresponding author. SLAC National Accelerator Laboratory, Menlo Park, CA, 94025, USA.
E-mail address: phsun@stanford.edu (P. Sun).

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defects act as trap sites for the diffusing D and T atoms, leading to the accumulation of these fusion fuel atoms within the wall material.

As recently shown [3], there are stringent limits on the allowed trapping probability of T atoms within the wall and divertor materials, and thus a deep, physics-based understanding of the trapping mechanisms is needed in order to reliably predict T retention in the designs of future devices such as DEMO. The development of such an understanding requires knowledge of the type, size, and density of the traps, along with a characterization of the trapping energy and capacity for each type of trap. The latter can be computed using, *e.g.*, density-functional theory (DFT) models [4,5]. The physical mechanisms involved in defect formation are rather complicated and extend through multiple time scales [2], making it difficult to predict the precise defect statistics using computer simulations. Hence, a statistical measurement of defect density and type can help benchmark simulation models and reveal the mechanism of property changes in the material. In addition, the development of these damage-induced defects may cause significant changes in the thermomechanical properties of the material [6–8] that can influence the subsequent engineering performance of the material. Again, knowledge of the defect statistics is needed to understand the resulting thermomechanical property evolution. It is therefore important to study the defect microstructure formed in displacement damaged tungsten found in the wall and divertor regions of fusion devices.

There have been a number of defect characterization studies on ion-irradiated tungsten, primarily using transmission electron microscopy (TEM). Among them are early works by Häussermann [9–11] and Jäger [12] using 20–70 keV Au^+ ions to irradiate W thin films at room temperature. They reported mostly vacancy-type dislocation loops with Burgers vector $\boldsymbol{b} = a_0/2\langle 111\rangle$. More recently, Yi et al. [13–15] did a series of systematic studies using 150 keV or 2 MeV W self-ions for irradiation on polycrystalline tungsten samples at a wide range of temperatures. Their work revealed details about the microstructure evolution with increasing dose and the effects of irradiation temperature on defect density and distribution. These studies have shown that TEM is a useful and direct technique to obtain detailed information about individual defects; statistical information can also be obtained by simply counting the defects. However, a main drawback is the requirement for a very thin sample ($\lesssim$ 100 nm sample thickness) which may lead to surface effects and, if sample preparation is done post-irradiation, it may affect existing defects and/or create additional defects.

Meanwhile, asymptotic X-ray diffuse scattering around Bragg peaks has been developed and verified as a technique to study the statistical distribution of defect clusters [16]. By utilizing the "single defect approximation" (see Section 2.1), statistical information on the type and concentration of defects can be obtained from a single X-ray diffuse scattering pattern. Past works [17,18] have shown that results from this method agree with TEM in the size range to which both methods are sensitive (usually 2–20 nm in diameter). Compared to TEM, X-ray measurements are non-destructive and requires no extra sample preparation. Moreover, X-ray diffuse scattering has the advantage of being able to probe into a bulk sample (a few micrometers or more in depth) and sample a large area (up to mm^2), whereas TEM can only image thin foils with a typical field of view of μm^2 or less. In addition, it is much easier to separate vacancy- from interstitial-type defects using X-ray diffuse scattering due to the opposite signs of lattice strain from these two types of defects (see Section 2).

Therefore, in this work, we use X-ray diffuse scattering around Bragg peaks to study defect clusters in bulk single-crystal tungsten. The samples are irradiated with copper ions at room temperature with different doses: 0.2, 0.6, 2, and 5 displacements per atom (DPA). For each sample, we are able to obtain the concentration of dislocation loops with radii between 5 and 80 Å and their distribution with respect to size and type (vacancy/interstitial). By comparison across samples, we also observe trends with increasing radiation dose, such as an increase in vacancy loop size.

2. Theory

2.1. Single defect approximation

The theory of X-ray diffuse scattering from defects near Bragg reflections has been developed by Dederichs [19,20] and Larson [16]. Here we summarize the main results to be used in our data analysis. The diffuse scattering differential cross-section per solid angle Ω is given by

$$\left[\frac{d\sigma(\boldsymbol{K})}{d\Omega}\right]_{\text{diffuse}} = \sum_{m,n} r_e^2 f_m f_n \left[\left\langle e^{i\boldsymbol{K}\cdot(\boldsymbol{r}_m-\boldsymbol{r}_n)}\right\rangle - \left\langle e^{i\boldsymbol{K}\cdot\boldsymbol{r}_m}\right\rangle\left\langle e^{-i\boldsymbol{K}\cdot\boldsymbol{r}_n}\right\rangle\right], \tag{1}$$

where m and n are indices running over all atoms in the crystal, r_e is the classical electron radius, $\boldsymbol{K}$ is the scattering vector, f_m is the form factor of the m^{th} atom, and $\boldsymbol{r}_m$ is the position of the m^{th} atom. Angular brackets denote ensemble averages that are carried out in practice by volume averaging. We make the following assumptions: i) the concentration of defect c, defined as the number of defect clusters per atom, is much less than 1; ii) the defects are well separated so that non-linear effects due to interaction between two clusters can be ignored; iii) the defects are randomly distributed, and ergodicity is satisfied by a sufficiently large sample volume. The first assumption appears to be self-consistent in the current study; *i.e.*, the total defect concentration is much less than 1 (see Section 4). There are cases when the second and third assumptions do not hold true, for instance with the formation of loop strings at elevated temperature [14]. However, we do not expect such phenomenon to happen for the majority of the defect population, especially when defect density is low and bombardment is done at room temperature. Hence to a good approximation we take all assumptions to be true, in which case Eq. (1) simplifies to [20]

$$\left[\frac{d\sigma(\boldsymbol{K})}{d\Omega}\right]_{\text{diffuse}} \approx \sum_j c^{(j)} N \frac{d\sigma_{\text{d}}^{(j)}(\boldsymbol{K})}{d\Omega}, \tag{2}$$

where N is the total number of atoms, $c^{(j)}$ is the concentration (measured in number of defects per unit volume normalized to the lattice atom density) for defect type j, and $d\sigma_{\text{d}}^{(j)}(\boldsymbol{K})/d\Omega$ is now the diffuse scattering cross-section for a single defect of type j in an infinite crystal [16,20]:

$$\frac{d\sigma_{\text{d}}^{(j)}(\boldsymbol{K})}{d\Omega} = \left| \sum_{m\in\mathscr{D}^{(j)}} r_e f_m e^{i\boldsymbol{K}\cdot\boldsymbol{r}_m^{(j)}} + \sum_{n\notin\mathscr{D}^{(j)}} r_e f_n e^{i\boldsymbol{q}\cdot\boldsymbol{r}_n^0}\left(e^{i\boldsymbol{K}\cdot\boldsymbol{s}_n^{(j)}}-1\right)\right|^2. \tag{3}$$

Here $\mathscr{D}^{(j)}$ denotes the set of atoms inside the defect (for vacancy-type defects, we take f_m to be negative). For atoms surrounding the defect, we have decomposed the position of the n^{th} atom $\boldsymbol{r}_n^{(j)}$ into a regular part $\boldsymbol{r}_n^0$ which refers to the average expanded lattice