Experimental observation of the number of visible defects produced in individual primary damage cascades in irradiated tungsten

Enquiries about copyright and reproduction should in the first instance be addressed to the UKAEA Publications Officer, Culham Science Centre, Building K1/0/83 Abingdon, Oxfordshire, OX14 3DB, UK. The United Kingdom Atomic Energy Authority is the copyright holder.
Experimental observation of the number of visible defects produced in individual primary damage cascades in irradiated tungsten

D.R. Mason,¹ X. Yi,² A.E. Sand³ and S.L. Dudarev¹

¹CCFE, UK Atomic Energy Authority, Culham Science Centre, Abingdon, Oxfordshire OX14 3DB, United Kingdom
²School of Materials Science and Engineering, University of Science and Technology Beijing, 100083, Beijing, China
³Department of Physics, University of Helsinki, P.O. Box 43, FI-00014, Helsinki, Finland

This is a preprint of a paper submitted for publication in Europhysics Letters
Experimental observation of the number of visible defects produced in individual primary damage cascades in irradiated tungsten

D.R. Mason1, X. Yi2, A.E. Sand3, S.L. Dudarev1

1 CCFE, UK Atomic Energy Authority, Culham Science Centre, Abingdon, Oxfordshire OX14 3DB, United Kingdom
2 School of Materials Science and Engineering, University of Science and Technology Beijing, 100083, Beijing, China
3 Department of Physics, University of Helsinki, P.O. Box 43, FI-00014, Helsinki, Finland

PACS 61.72.J– Point defects and defect clusters
PACS 61.80.Az – Theory and models of radiation effects
PACS 68.37.Lp – Transmission electron microscopy (TEM)

Abstract – We present a new analysis of nanoscale lattice defects observed after low-dose in situ self-ion irradiation of tungsten foils at cryogenic temperature. For decades, defect counts and size-frequency histograms have been the standard form of presenting a quantitative analysis of the nanoscale “black-dot” damage typical of such irradiations. Here we demonstrate a new statistical technique for generating a probability distribution for the number of defects produced in a single cascade. We show that while an average of fewer than one defect is observed per incident ion, the number of cascades with two or more visible defects produced is significant.

Introduction. – In situ irradiation of TEM-transparent foils offers great control over damage production and thermal history, and becomes a very valuable tool for nuclear materials research when we can extract data comparable to our simulations of physical radiation damage processes. For decades, quantitative analyses of irradiation damage in the form of nanoscale defects as been presented as defect counts and size-frequency histograms [1–3], but the position correlation of the defects has been under-utilised, due to the time-consuming nature of identifying a sufficient number of spots by eye to make such an analysis practical. The importance of this data, present in micrographs but unexploited, has recently become clearer. Primary damage cascades are inherently random processes, and their subsequent microstructural evolution may well be dominated by rare events [4–7]: the evolution of point defects and small clusters will be strongly skewed if a large dislocation loop is also generated during the heat spike phase [8], and if multiple large loops are in close proximity then their elastic interaction can lead to self-trapping [9].

Recently we have developed techniques [10, 11] for automating the analysis of black-dot damage, which produces a reproducible list of spot positions and sizes in a few minutes. We have been able to verify that primary damage cascade events in ultra-high purity tungsten foil produce a power-law size-frequency distribution of defects [10], and have accounted for observed deviations from power-law behaviour due to subcascade branching [12]. We have also been able to show, from analysis of the pairwise radial distribution function, that the characteristic size of individual cascades is of order one nanometer [11]. In this letter we perform a new type of analysis, to find the number of visible defects produced in a single cascade.

In situ TEM experiments. – Experimental data for the count of visible defects per cascade is generated from in situ self-ion irradiations of high purity tungsten foils, performed at cryogenic temperature at the IVEM-Tandem Facility at Argonne National Laboratory. It is known that the collapse of displacement cascades in self-ion irradiated tungsten produces large, nanometre-scaled (and therefore TEM visible) dislocation loops [9, 10]. We perform experiments at cryogenic temperature (30K), where the mobility of irradiation-induced defects is reduced, though we acknowledge that Brownian motion of defects due to quantum fluctuations of atomic positions associated with their zero point motion will still be present [13] and so loop loss to the surface can still occur.

3mm discs were cut from ultra-high purity tungsten
sheets (supplied by Plansee with nominal > 99.996 wt% purity). These were then heat-treated in vacuum at 1673 K for 20 hours, before being electropolished to electron transparency thickness (≤100nm). See refs [9,14] for full technical details of the sample preparation. The sample was irradiated in situ, at an incidence angle 15° off normal, up to a fluence of 1.25 × 10^{20}W^+/{m}^2.

(001) grains were selected for analysis, imaged in weak-beam dark-field conditions, using (g =200, 4.25g; g =200, 4.75g; g =200, 5.25g; g =110, 5.25g; g =110, 7.25g; and g =110, 7.75g). Regions were superimposed using the convergent weak beam technique [15], and analysed using the automated technique described in [10,11]. We present re-Analysis. It is important to note that while many dislocation loops can be counted, appearing on the micrographs as spots [18], their proximity alone is insufficient to confidently state that a pair originates from a single cascade. A statistical method is required to find the most likely distribution function for the number of visible defect clusters per cascade event.

We want to compute the probability of a cascade generating exactly m spots on the micrograph, which we denote \( P_{\text{cluster}}(m) \). Start by considering the probability of finding k spots in an observed area A. This probability must be the sum over all possible combinations of having a number \( k_1 \) one-spot cascades, \( k_2 \) two-spot cascades and so on.

\[
P_{\text{total}}(k; A) = \sum_{k_1, k_2,...} \delta_{k_1+2k_2+...+k} p(k_1, k_2, \ldots; A),
\]

where \( p(k_1, k_2, \ldots; A) = p(k_1; A)p(k_2; A) \ldots \).

\( p(k; A) \) is the probability distribution of observing \( k \) m-spot cascades in an area A. This probability is expected to be Poisson distributed as their generation is independent. The average count of m-spot cascades in an area A is \( \lambda_m = \rho P_{\text{cluster}}(m) \), where \( \rho \) is the irradiation density (incident ions per unit area).

\[
p(k; A) = P_{\lambda_m}(k) = \frac{(\lambda_m)^k e^{-\lambda_m}}{k!},
\]

where \( \lambda_m = P_{\text{cluster}}(m) \). Similarly the expected number of m-tuples of spots in area A is a sum over all possible combinations of m spots:

\[
\langle n(m; A) \rangle = \sum_k m! P_{\text{total}}(k; A) = \sum_k \sum_{k_1,k_2,...} \delta_{k_1+2k_2+...+k} P_{\lambda_1}(k_1)P_{\lambda_2}(k_2) \ldots
\]

This can be measured from the micrographs as follows. For each spot in the micrograph we find each possible set of m spots which includes it. We construct the minimum
The number of visible defects produced in individual primary damage cascades

Fig. 1: Left: TEM micrograph of in situ irradiation of 150keV self-ion irradiation of UHP tungsten foil at 30K. WBDF imaging shown with highly exaggerated contrast. Right: Analysis of the position and size of defect clusters. Colours denote defect cluster diameter. The proximity of spots can not be used to determine if they originated in one cascade. Rather our analysis proceeds by finding minimum enclosing circles containing m spots, and fitting the distribution. A circle with diameter 9.2nm, containing m = 4 spots is indicated.

<table>
<thead>
<tr>
<th>t*</th>
<th>α</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.1</td>
<td>0.15</td>
</tr>
<tr>
<td>6.3</td>
<td>0.025</td>
</tr>
<tr>
<td>12.7</td>
<td>0.001</td>
</tr>
</tbody>
</table>

Table 1: Critical values of $t^*$ used to find a significance level $\alpha$, with the settings used for the analysis of these images. A significance value $\alpha$ is the probability a type-I error - the natural background variation being (erroneously) identified as a spot.

<table>
<thead>
<tr>
<th>Ion energy (keV)</th>
<th>50</th>
<th>150</th>
<th>400</th>
</tr>
</thead>
<tbody>
<tr>
<td>regions studied</td>
<td>4</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>total area (nm²)</td>
<td>1.71e+07</td>
<td>2.96e+07</td>
<td>3.05e+07</td>
</tr>
<tr>
<td>incident ions</td>
<td>21300</td>
<td>37000</td>
<td>36100</td>
</tr>
<tr>
<td>visible clusters per cascade</td>
<td>0.15 [0.06:0.22]</td>
<td>0.14 [0.08:0.21]</td>
<td>0.21 [0.16:0.27]</td>
</tr>
<tr>
<td>ion depth (nm)</td>
<td>6.9</td>
<td>13.7</td>
<td>30.6</td>
</tr>
<tr>
<td>inter-cascade length (nm)</td>
<td>8.2</td>
<td>10.5</td>
<td>13.4</td>
</tr>
</tbody>
</table>

Table 2: Results for the analysis of primary cascades in TEM micrographs. The depth of the damage region $L$ is computed using SRIM [16]. We have computed a typical inter-cascade length assuming uniform cascade generation within this depth, combined with the ion fluence $1.25 \times 10^{16}$ W^+m^{-2}. The number of visible clusters per cascade has three values- we report the significance level $\alpha = 0.025$, with the bracketed region the interval $[\alpha = 0.001, \alpha = 0.15]$.

enclosing circle (MEC) around the set, defined to enclose the centres of the spots as determined by analysis of the micrograph. Write $N(m; A)$ as the number of minimum enclosing circles containing at least $m$ spots with area less than or equal to $A$. Note that $N(1, A) = N(1, 0)$ is the total count of spots in the micrograph. We can then best fit $P_{\text{clust}}(m)$ to match the (theoretical) $\langle n(m; A) \rangle$ to (experimental) $N(m; A)$, normalised to reproduce the total count of spots.

We present the results for $N(m; A)$ in figure 3 using minimum enclosing circles with diameter less than or equal to $d = 8\text{nm}$. We choose this characteristic diameter based on our recent detailed analysis of the spatial distribution of primary damage [11], though the results are in fact insensitive to size between $d = 8\text{nm}$ and $d = 16\text{nm}$. The $t^*$ thresholds for the identification of smaller, fainter spots give error bars for this calculation. The lower threshold we consider ($\alpha = 0.15$ ) gives the most spots. This level also gives the upper limit of the number visible spots $m$ per MEC. This proves that the faintest spots we are recording are clustered, and so suggests that this low threshold is capturing faint dislocation loops produced in cascades.

We present fits for the number of defect clusters produced per cascade, $P_{\text{clust}}(m)$ in figure 4.

Without any prior assumptions, except for no cascade overlap, we find that while very few cascades produce vis-
Fig. 2: The experimentally observed size-frequency histogram for the three ion implantation energies considered. The shading indicates the count of spots with intensities over significance levels $\alpha = 0.001$ (black), through 0.025 (mid-grey) to 0.15 (light grey). Presenting the data in this manner makes it clear that smaller, and therefore fainter loops are more difficult to confidently identify on the micrograph.

Fig. 3: The experimentally observed count, $N(m; A)$, for the $m$-tuples of spots which may be contained in a minimum enclosing circle (MEC) with diameter less than or equal to $d = 8$ nm (see text). Error bars denote counts of spots with intensities over different critical thresholds, with significance levels $\alpha = 0.15$ (higher value of error bar) to $\alpha = 0.001$, with the symbol at significance level $\alpha = 0.025$. The shaded region indicates a best fit for this observed count, assuming that individual cascades are independently generated. Again the upper and lower bounds correspond to intensity threshold significance levels of $\alpha = 0.15$ and $\alpha = 0.001$.

Discussion and Conclusions. – An interesting question to ask is whether the distribution $P_{\text{clust}}(m)$ itself fits a simple analytical form. The simplest possible model to propose is that $P_{\text{clust}}(m)$ is Poisson distributed. This would suggest that large visible defects are produced independently of each other within a cascade. We could argue that the kinetic energy of the incident ion is nearly all lost to phonons, with only a small fraction remaining as the excess potential energy of the residual lattice defects, so the Poisson process is within energy space. The difficulty with this model is that any subsequent evolution of the microstructure, be it loss of interstitial clusters to the surface or aggregation/recombination of defects would bring the observed distribution away from Poisson. Recent theoretical and experimental observations [9, 11, 12], suggest a dominant mechanism for loop retention in a thin foil irradiation experiment like this one is mutual elastic self-trapping. Put simply, if only one large loop is generated in a cascade, there is little to prevent it being attracted to the surface and lost [19, 20]; if two or more are generated they can mutually self-trap [21]. We might therefore ex-
The number of visible defects produced in individual primary damage cascades

![Graphs showing the fitted distribution for the number of visible spots generated in individual primary damage cascades. Error bars denote counts of spots with intensities over different critical thresholds, with significance levels α = 0.15 (higher value of error bar) to α = 0.001, with the symbol at significance level α = 0.025. Only the assumption of independent (non-overlapping) individual cascades is used for this fitting. The shaded region indicates a possible alternative fit (using the same threshold intensities for sampling the spots), with the additional assumption that the number of dislocation loops produced in each cascade is Poisson distributed.](image)

Fig. 4: The main result of this letter, the fitted distribution for the number of visible spots generated in individual primary damage cascades. Error bars denote counts of spots with intensities over different critical thresholds, with significance levels α = 0.15 (higher value of error bar) to α = 0.001, with the symbol at significance level α = 0.025. Only the assumption of independent (non-overlapping) individual cascades is used for this fitting. The shaded region indicates a possible alternative fit (using the same threshold intensities for sampling the spots), with the additional assumption that the number of dislocation loops produced in each cascade is Poisson distributed.

pect to see fewer single-spot-cascades than were actually generated. Fitting \( P_{\text{clus}}(m) = \text{Po}\_f(m) \) for a single value \( f \) to the observed count of \( m \)-tuples per MEC, ie \( N(m;\lambda) \), gives the shaded region in figure 4. This appears to be a good approximation for the best fit we were able to produce. We should stress that coincidence of these results should not imply causality - we have not proved here that visible defects are produced independently within individual cascades, only that the experimental evidence we have gathered is not inconsistent with this model.

We conclude that in these in-situ ion irradiations, the incident ions initiate cascades, and in the heat spike phase nanoscale dislocation loops can be generated. While most cascades do not produce any visible defects, some produce loops large enough to be seen as spots in the TEM (\( \geq 2 \) nm in our imaging conditions). There is no evidence for subsequent growth at cryogenic temperatures and very low fluence. In this paper we have shown that where large loops are generated, they do not only appear as isolated singletons, but can appear as pairs or triplets.

This work proves that rare events - the simultaneous production of multiple large loops within a single cascade - can not only be observed, but are likely to be important stable nucleation sites for subsequent microstructural evolution in irradiated materials.

Acknowledgements. – The authors would like to thank Jack Haley, Oxford University, for many helpful discussions about the presentation of size-frequency histograms.

This work has been carried out within the framework of the EUROfusion Consortium and has received funding from the Euratom research and training programme 2014-2018 under grant agreement No. 633053, and was part-funded by the Research Councils UK Energy Programme [grant number EP/P012450/1]. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

The experiments of in situ ion irradiations at liquid helium temperature were carried out at Argonne National Laboratory, using the IVEM-Tandem Facility. We thank to Dr Marquis Kirk, Pete Bald and Edward Ryan for their help with the irradiations. This work was supported by a U.S. Department of Energy Facility funded by the DOE Office of Nuclear Energy, operated under Contract No. DE-AC02-06CH11357 by U. Chicago Argonne, LLC.

REFERENCES


