The spatial distribution of self-interstitial atoms around depleted zones in tungsten ion-irradiated at 10 K

By CHING-YEU WANG and DAVID N. SEIDMAN
Cornell University, Barrett Hall, Department of Materials Science and Engineering and the Materials Science Center, Ithaca, New York 14853, U.S.A.

[Received 7 July 1980 and accepted 14 November 1980]

ABSTRACT

The three-dimensional spatial distribution of self-interstitial atoms (SIAs) around depleted zones (DDs) in ion-irradiated tungsten was determined by field-ion microscopy (FIM). High-purity, four-cast, non-replicated, single-crystal tungsten FIM specimens were irradiated in situ with 5 MeV Si"" ions along the [111] direction, at 10 K, and examined at the same temperature by the pulse-field-evaporation technique. At 10 K, the SIAs are completely immobile. The distances were measured along the short-packed crystallographic directions—that is, in the (100), (110) and (111) directions—between each SIA and the DDs. Distance measurements were also made between each SIA and the irradiated surface of the specimen. The net of distances employed for analysis was superimposed on the minimum measured distances. The histograms of the distribution function represent an upper bound (denoted by $\Delta_{\text{max}}$) to the actual configuration distances. For the 5 MeV Si"" ion-irradiated specimen, a total of 97 SIAs were counted, and $\Delta_{\text{max}} = 175 \pm 25$ A. The $\Delta$ values for $(\Delta_{\text{max}})$ represent the standard deviation. In the case of the 18 MeV Ar"" ion-irradiated specimen, the quantity $(\Delta_{\text{max}})$ is $155 \pm 20$ A. 12 SIAs were detected. A comparison of $\Delta_{\text{max}}$ values was obtained by combining the former measurements (Huang et al., 1979a and Seidman et al., 1979) with the present results to obtain $\Delta_{\text{max}} = 160 \pm 20$ A. It is suggested that this value may represent an overestimate of the mean range of replacement collision sequences in tungsten. Nevertheless, the results constitute very direct evidence for the existence of DDs in tungsten.

§1. INTRODUCTION

For over two decades the subject of focused collision sequences (FCSs) and replacement collision sequences (RCSs), in the field of radiation damage, has evolved a great deal of theoretical and experimental research. The beginnings of this subject date from the seminal paper of Glazebrook (1967), in which he introduced the concept of an RCS or focus in an irradiated crystal lattice. The existence of RCSs provides a mechanism for the efficient transport of momentum along short-packed crystallographic directions without the permanent displacement of lattice atoms. Shortly thereafter Vaneyard and co-workers (Gibson, Golsan, Kitgum and Vineyard 1967, Vaneyard and Eglington 1984, Kegnass, Vaneyard and Shinnosuke 1985), in a series of epoch-making papers, employed
the molecular-dynamics computer technique to simulate low-energy radiation damage events. The computer experiments carried out by Vasserman and co-workers showed the existence of FCSs and a new type of event that is now called an RCS. RCSs were observed in both the face-centred (copper) and body-centred (zirconium) cubic lattice. As it involves the transport of mass as well as momentum along characteristic crystal lattice directions, as a result of this mechanism it is possible to dynamically produce a self-interstitial atom (SIA) a number of interstitials distances away from its vacancy. Seeger (1958, 1962) also postulated the concept of a dynamic crowding—RCS in our vernacular—as a means of separating the SIA from its vacancies in a displacement spike (Seeger 1954). Both FCSs and RCSs are low-energy phenomena (typically less than 1 keV of transferred energy) and they have been discussed extensively as one of the important factors that determine the initial spatial arrangement of vacancies and SIA in irradiated crystals (see, for example, Thorel 1969, Nielson 1968); that is, the spatial arrangement prior to the start of extensive thermally activated migration of point defects. Unfortunately, the direct experimental measurement of the mean range and range distribution of either FCSs or RCSs has proved to be a major scientific problem. For example, different types of experiments—on gold—has been interpreted as indicating a mean range of less than 30 Å (Ecker et al. 1970) and greater than 2000 Å (Seeger 1970). Various aspects of the copper experimental situation have been reviewed by Vasservis (1970), Birk-Kirk and Scott (1975) and Thompson (1978). Also, recently, Tenenbaum and Doan (1977) and Tenenbaum (1978) have employed the molecular-dynamics computer technique to calculate the effects of lattice vibrations on RCSs. Despite all this time and effort, the subject of RCS and its range distribution has remained extremely controversial.

Tungsten is an interesting element to study in the search for SIA produced by RCSs, as it is a heavy element with a reasonably large atomic radius; therefore, one would expect focusing effects to be important in the lattice (see, for example, Thompson 1969, Nielson 1968). The present report presents new results on the spatial distribution of SIA around depleted zones (DZs) created as a result of in situ irradiation of tungsten field-ion specimens at 10 K, with 30 keV Os or 18 keV Au ions. At 10 K, in tungsten, we have demonstrated that SIA are completely immobile (Kallmen, Storitz and Seidman 1971, a; Seidman, Wilson and Nielson 1975 a, b; Wilson and Seidman 1977, Wilson and Seidman 1980). The experimental approach was similar to the one employed in an earlier piece of research (Bearden, Scandro and Seidman 1971), although the FIM techniques have been refined considerably since that publication (Seidman 1978). The basic procedure was to displace a specimen at 10 K, after an in situ irradiation, on an atom-by-atom basis to search for both the DZs and SIA. Thousands of FIM images were recorded on 35 mm cine film, which were subsequently analysed.

+ We have used this term—in preference to 'displacement spike'—to refer to the region of high vacancy concentration after all the deposited energy has been dissipated.

### 2. EXPERIMENTAL DETAILS

#### 2.1. Specimen chemistry and preparation

Four-pass, zone-refined single crystal rods 1.5 to 6.5 mm in diameter and 26 cm long were prepared by the electron-beam zone-melting technique. They were first reduced to a 0.5 mm diameter rod by electroplating at 20 V d.c. in a solution of 20 g NaOH in 1 l of a 60–40 mixture of glycerol and water, with air bubbled through the electrolyte. The drawn rods were then polished and sectioned into 2 cm lengths (0.5 mm in diameter) in a 1N NaOH electrolyte at 9 V d.c. A section of the specimen was mounted in a specimen holder and polished to a sharply focused FIM tip (by the a.c. drop-off technique (Muller and Thong 1969). The specimen was immersed vertically into a layer of 1N NaOH aqueous solution which was floated on top of CuO. After the specimen and a stainless-steel counter electrode until a highly seeded region was produced at the interface between the NaOH and the CuO. The specimen was next raised slightly and the rod was allowed to propagate inward until the lower section of the wire dropped off. At this point the specimen had a thin and long taper. It was polished with the procedure described above, but before the lower section of the wire dropped off the voltage was switched to a ~ 3 V a.c. pulse controlled by a programmed push-button switch. The pulse was terminated when the upper section was observed to drop into the solution. A specimen produced in this a.c. drop-off technique imaged at a potential of 3 to 4 keV in the IMF, this corresponded to a tip radius of ~ 100 Å. The initial end-form of the metastabilized tip was extremely rough on an atomic scale. An atomic smooth end-form was obtained by combining d.c. and pulse-field etching at 75 K.

#### 2.2. The irradiation procedure

After a specimen had been field-evaporated to a tip radius of 200 to the specimen was cooled to 12 K and brought to an atomically smooth low-
temperature end-form by the pulse-field-evaporation technique. The specimen was irradiated in situ at a background pressure of 2 x 10⁻¹⁰ Torr—in the absence of the electric field and imaging gas—with a magnetically analysed 30 keV Cr⁺ ion beam to a total dose of 4 x 10¹⁵ ions cm⁻². The flux of ions at the specimen was ~ 1 x 10¹⁵ ions cm⁻² s⁻¹. A second specimen was irradiated with a magnetically analysed 18 keV Ar⁺ ion beam to a total dose of 5 x 10¹⁴ ions cm⁻²; the flux of ions at the specimen was ~ 2 x 10¹⁵ ions cm⁻² s⁻¹. In both cases the time required to form an atomic monolayer was always greater than the irradiation period. After a 10⁻⁵ s irradiation each specimen was re-imaged, employing helium as the imaging gas, at ~ 10⁻⁵ Torr. Each specimen was dissected atom by atom employing the pulse-field-evaporation technique (Seidman 1973, 1974, 1976, Seidman et al. 1975 a, b).

For details regarding temperature control and measurement, the sputtered-metal ion-source and the irradiation facility, see Seidman, Scanlan, Styris and Bohlen (1969), Seidman and Scanlan (1971), Scanlan et al. (1971 a, b), Petroff and Seidman (1973) and Wilson and Seidman (1975).

2.3. Data recording system and cine-film analyser

An FIM image was recorded photographically after a field-evaporated pulse had been applied; the pulse height and duration were adjusted within one atom, on average, was removed per pulse on the (222) plane. The FIM images were recorded on 35 mm cine film employing an Automax camera equipped with a 1000 f.l.t. film chamber. The 35 mm cine film was analysed with a Vanguard motion analyser which was interfaced to a Houston Osmograph 200 x-y recorder; see our earlier publications for further details (Scanlan, Styris, Seidman and Ast 1969, Beaven et al. 1972, Wilson and Seidman 1973, We and Seidman 1978).

§3. CONTROL EXPERIMENTS

Control experiments were performed on unirradiated tungsten specimens to detect possible artifact vacancies and SIAs. The term "artifact vacancy" or "SIA" implied the presence of a vacancy or an SIA which was not the result of an irradiation. A vacancy appeared as a dark spot at the position of an atomic site. It has been shown that an SIA can produce a contrast pattern (Seidman and Lie 1972) consisting of (a) a bright spot on an extra-bright spot and (b) a vacant lattice site.

The criteria employed for counting both atomic sites and vacancies were as follows: (1) for small net planes (< 20 atoms) we counted both atomic sites and vacancies within the first ring of atoms (see fig. 1) immediately after the last atom of the preceding plane had been field-evaporated; and (2) for large net planes (> 20 atoms) we counted both atomic sites and vacancies within the first ring when the first two outer rings of atoms become visible, even though several atoms of the preceding plane were retained. Figure 1 shows a schematic diagram of an (84) net plane.

† At 10 K we have demonstrated that SIAs are immobile (Scanlan et al. 1971 a, b), Seidman et al. 1975 a, b, Wilson and Seidman 1975, Wilson et al. 1975.

In a total number of ~ 1 x 10⁷ atomic sites counted in two well-annealed tungsten specimens, Park (1975) found only one f-dark spot contrast effect; a second spot is a single dark-spot that appears suddenly within a well-resolved net plane during observation and remains there until all the atoms of that plane are evaporated. This f-dark spot contrast effect, however, was not counted as a vacancy in our work. The origin of this contrast effect was most likely a result of interaction of the imaging gas with the atoms in the surface of the specimen.
Artifact vacancy concentrations detected in different vicinal planes near the (411)
pole of a irradiated pure tungsten specimen.

<table>
<thead>
<tr>
<th>Plane</th>
<th>Number of atomic sites</th>
<th>Number of artifact vacancies</th>
<th>Artifact vacancy concentration (at.%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(411)</td>
<td>68,582</td>
<td>28</td>
<td>$3.5 \times 10^{-4}$</td>
</tr>
<tr>
<td>(212)</td>
<td>109,655</td>
<td>3</td>
<td>$2.7 \times 10^{-3}$</td>
</tr>
<tr>
<td>622</td>
<td>61,615</td>
<td>337</td>
<td>$8.6 \times 10^{-4}$</td>
</tr>
<tr>
<td>631</td>
<td>66,064</td>
<td>9</td>
<td>$9.1 \times 10^{-3}$</td>
</tr>
<tr>
<td>721</td>
<td>71,443</td>
<td>42</td>
<td>$5.0 \times 10^{-4}$</td>
</tr>
<tr>
<td>732</td>
<td>60,497</td>
<td>120</td>
<td>$1.0 \times 10^{-3}$</td>
</tr>
<tr>
<td>831</td>
<td>104,674</td>
<td>8</td>
<td>$7.0 \times 10^{-4}$</td>
</tr>
<tr>
<td>842</td>
<td>108,793</td>
<td>171</td>
<td>$1.6 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

counted for each plane are listed in the table. In most cases the artifact vacancy concentration is between $1 \times 10^{-6}$ and $1 \times 10^{-2}$ at.%, with the exception of the [622] planes. In addition, no contrast patterns of SIA were detected among a total of $\sim 7 \times 10^{6}$ atomic sites counted in these studies; hence, the artifact SIA concentration for the planes near the (411) pole is $< 1.4 \times 10^{-2}$ at.%. In conclusion, the results presented in this paper were not affected by the presence of artifact SIA or vacancies.

§ 4. Results

4.1. The 30 keV Cr$^+$ ion irradiation

In one specimen which had been irradiated with 30 keV Cr$^+$ ions, we found five depleted zones (DZs) and a total of 97 SIA in the layers surrounding these DZs; the SIA concentration is $\sim 9 \times 10^{-3}$. Figure 2 (a) shows a partial [211] standard stereographic projection which shows a top view of the specimen; this specimen had a [211] orientation and the ion beam was parallel to the [741] direction. Since the ion beam was parallel to this very-high-index crystallographic direction, the probability of channelling was minimized (see, for example, Whitten 1973). A plane (or planes) in which a depleted zone was detected is also indicated. DZs was found near the [222] plane of the specimen. A cross-sectional side-view of the specimen is shown in fig. 2 (b). The distance between any two DZs is indicated by a, and so on, the distances range from 160 to 370 Å.

Figure 3 exhibits a pulse dexterity sequence through a typical SIA contrast pattern detected in this specimen. This SIA contrast pattern extended through six (732) planes and a total of 140 frames of cinefilm.

4 In only one out of the four tungsten specimens which had been irradiated with 30 keV Cr$^+$ ions did we find DZs which were capable of being mapped with detail; they are denoted as DZa and DZb. The results on the vacancy type of these depleted zones will be reported separately (Wei, Current and Seidman 1978).

5 This corresponds to a distance of 241 Å. The crystallography of the plane is given in Nicholas's atlas (Nicholas 1966).
A series of 12 FIM micrographs out of 58 recorded during the atom-by-atom dissection of nine (221) planes. The contrast patterns of the SIA extends through six (221) planes. The positions of the atoms and the vacancies in the (221) planes are indicated schematically below each micrograph; the symbols employed are the same as in fig. 3.
Figure 4 shows a second example of the contrast pattern of an SIA. This SIA contrast pattern extends through six (521) planes and a total of 581 frames of cine film were involved in this analysis; only 12 frames of film are shown in fig. 4. Layer numbers 4, 5, 6 and 7 (see frames 239, 306, 336 and 403) contain six vacancies. An extra bright-spot contrast effect is also seen in a non-lattice site in layer 5 (see frames 232, 257 and 340). Therefore, an SIA can produce a contrast pattern consisting of: (1) a bright spot, (2) an extra-bright spot and (3) a vacant lattice site (Seidman and Lee 1972). This type of contrast pattern was only observed in the irradiated specimens and therefore was not caused by artifacts (see §3).

A computer-generated isometric drawing of the 30 keV Cr⁺ irradiated specimen is shown in fig. 5. The five open ellipses represent the depleted zones (DZ5a to DZ5e) and the solid black circles indicate the SIA. The mathematical details of the mapping procedure are given in the Appendix.

It is noted (see fig. 5) that the DZs are located mainly on the incident beam side of the specimen, whereas a large fraction, of the total number of SIA observed, was detected on the side of the specimen shielded from the Cr⁺ ion beam. The clear separation between the DZs and SIA is evident in fig. 5.

The distances were measured—along the close-packed directions in the (100), (110) and (111) directions—between each SIA and the nearest (111) plane. These values were then compared to those of Nicholas (1965) for the same specimen, and it was found that the distribution of SIA was consistent with the expected distribution.

Figure 6 is a histogram of the number of SIA per 50 Å bin versus $R_{max}$ for the tungsten specimen which was irradiated with 30 keV Cr⁺ ions at 10 K. The open ellipses indicate DZs and the solid circles indicate the SIA. The error bars represent the standard deviation ($\sigma$). It is emphasized very strongly that the histogram may not represent the actual distribution of SIA; however, it does provide strong evidence for the existence of SIA.
4.2. The 18 keV Au\textsuperscript{+} irradiation

In a specimen which had been irradiated with 18 keV Au\textsuperscript{+} ions we detected a total of 33 SIAs: all the depleted zones produced were located within ≤ 30 Å of the irradiated surface. The value of 18 keV and the heavy mass (196.967 a.m.u.) of the projectile ion were chosen so that all the DZs were created, or slightly below, the irradiated surface. The ion beam was parallel to the very-high-index [714] direction, so once again the probability that the incident ion beam became channelled was negligible (Whitton 1973). This spatial arrangement of DZs implied that the distance from an SIA to a DZ was almost equal to the distance from an SIA to the irradiated surface.

A computer generated isometric drawing of the specimen which had been irradiated with 18 keV Au\textsuperscript{+} ions is shown in fig. 7. The SIAs that were injected into the bulk of the specimen are indicated by the solid black circles; the DZs are denoted schematically by the open circles. The distances between each SIA and the surface of the specimen were measured along all the possible (100), (110) and (111) directions. In this case we also took the set of distances which corresponded to the minimum measured distances as $R_{\text{min}}$ (see § 4.1). Figure 8 presents a histogram of the number of SIAs per 50 Å bin versus $R_{\text{max}}$. The quantity $\langle R_{\text{max}} \rangle$ is 175 ± 15 Å, where the ± values are for one $\sigma_{\text{max}}$. This value is in good agreement with the value of 175 ± 110 Å measured for the 30 keV Cr\textsuperscript{+} ion irradiation.

\[ \langle R_{\text{max}} \rangle = 175 \pm 15 \text{ Å} \]

5.1. The number of SIAs detected

The total number of SIAs detected in both specimens was always less than the total number of vacancies measured. In the case of the 30 keV Cr\textsuperscript{+} ion irradiation we determined experimentally that DZs and DZ's contained 241 and 247 vacancies, respectively (Wei 1978, Wei et al. 1980). It was not possible to measure the number of vacancies contained within DZs to DZ's, but an average value of ~ 172 vacancies per DZ for 30 keV Cr\textsuperscript{+} ion irradiations was found to be a reasonable number (Wei 1978, Wei and Seidman 1979). Thus the fraction of SIAs detected was approximately ~ 0.1 of the total number of vacancies produced in the case of 30 keV Cr\textsuperscript{+} ion irradiation.

\[ \frac{\text{Number of SIAs detected}}{\text{Total number of vacancies}} = 0.1 \]

The most obvious physical reasons for the above discrepancy are as follows:

1. It was only possible to examine a small fraction (0.1 to 0.2) of the specimen.

2. The value was estimated from the total number of 18 keV Au\textsuperscript{+} ions and the total number of atoms contained within $V_{\text{tip}}$ examined. The quantity $V_{\text{tip}}$ is given by:

\[ V_{\text{tip}} = (2/3)(r_1^3 - r_2^3)(1 - \sin \alpha)^3/\sin \alpha, \]

where $r_1$ and $r_2$ are the initial and final radii of the tip and $\alpha$ is the shank angle of the tip.

\[ V_{\text{tip}} = (2/3)(r_1^3 - r_2^3)(1 - \sin \alpha)^3/\sin \alpha, \]

In this drawing the positions of the DZs are simply schematic for the detailed vacancy structure of the DZs see Wei et al. (1980).
the total tip volume ($V_{tip}$) in our search for SIAs, since only certain high-index planes were found to be satisfactory for detecting SIA contrast patterns (Beavan et al. 1971, Seidman and Wei 1972, Seidman 1973, Wei 1978). (2) A fraction of the RCSs was propagated away from the DZs towards the irradiated surface; the upper limit to this fraction is less than 0.5. If the average value of the RCS range is greater than the distance of a DZ from the irradiated surface then some unknown fraction of these SIAs would have been shot out of the specimen. Figure 5 shows that only a few SIAs were detected in the half of the specimen which was exposed to the ion beam. Thus this reason also appears to be a very plausible one. (3) If the average RCS range is greater than the distance from a DZ to the unirradiated surface, then many of the resulting SIAs could not have been detected. It is difficult to evaluate this possibility quantitatively since we do not know a priori the average RCS range and the parent distribution of ranges of the SIAs which are deposited at the end of the RCS chains.

Reasons (1) and (2) are sufficient to explain the fact that the fraction of SIAs detected was ~0.1 of the total number of vacancies produced. Thus there seems to be no compelling need to invoke reason number (3)—this point is discussed further in § 5.2.

5.2. The distribution of ranges of the replacement collision sequences

In §§ 4.1 and 4.2 we have presented distributions for the ranges of RCSs in tungsten FIM specimens which had been irradiated with 18 keV Ar+ or 30 keV Cs+ ions, along the [110] direction; the value of $\langle R_{max} \rangle$ for these distributions is 175 ± 110 Å. The data in figs. 6 and 8 were combined with our earlier data (Beavan et al. 1971) to produce the composite distribution exhibited in fig. 9: the value of $\langle R_{max} \rangle$ for this distribution is $\approx 160 \pm 120$ Å. To the best of our knowledge these are the only distributions (figs. 6, 8 and 9) of this type ever determined for RCSs.

The main problem with these range distributions is that we do not know the relationship between them and the parent standard distribution of RCS ranges. The problem is not simply a statistical one but rather a physical one because of the limitations of our sampling procedure [§ 4.1]. Thus, for example, if a large number of SIAs in the immediate vicinity of each DZ are to be detected, then our value of $\langle R_{max} \rangle$ would represent an overestimate of the mean range of RCSs. There is no plausible reason to believe that our $\langle R_{max} \rangle$ values underestimate the mean range of RCSs. The reason for this is that the fraction of SIAs per 50 Å bin (fig. 9) does not show any monotonicity with distance from the DZs, whereas one would expect the opposite behaviour if a large fraction of the RCSs produced had travelled appreciable distances—along the close-packed crystallographic directions—without losing a significant fraction of their energy before they had come to rest as SIAs. It is noted, in connection with the latter point, that in the simulation of low-energy radiation damage events by the molecular dynamics technique, no evidence was found for RCSs which travelled appreciable distances with very small energy losses to the lattice (Vineyard 1980, Erginoy et al. 1964, Tenenbaum 1978). Thus it is our opinion that the $\langle R_{max} \rangle$ values do not underestimate the mean range of RCSs, but that they may overestimate it. The present distributions demonstrate that there were some RCSs with an appreciable range but the exact relationship between the measured fractions (see fig. 9) and absolute fractions of the parent standard distribution is unknown.

To quantify the above points somewhat, concerning the ranges of RCSs, we used the crude first-order focusing model—developed originally by Nisen and Thompson (1961) and Nelson (1963)—to estimate the focusing energies $\langle E_{RCS} \rangle$ and RCS ranges along different close-packed crystallographic directions in the b.c.c. lattice. The model of Nelson and Thompson assumed a repulsive Born-Mayer potential of the form

$$V_{w.w}(r) = A \exp(-br),$$

where $a$ is the lattice parameter and $b = A / a$.
where \( r \) is the internuclear separation, \( A \) a constant and \( b \) the reciprocal of screening length; we used \( A \) and \( b \) equal to 54.181 eV and 3.50171 Å, respectively (Abrahamsen 1960). The expressions for \( E_{10}^{\text{SI}} \) are (Nelson 1965)

\[
E_{10}^{111} = 2.4 \exp \left( -\frac{\sqrt{2}}{a_0 b} \right)
\]

(in a \{110\}-type plane)

\[
E_{10}^{110} = \frac{(\sqrt{2} a_0 b)^2}{A \exp \left( -\frac{\sqrt{2}}{a_0 b} \right)}
\]

where \( E_{10}^{111} \) and \( E_{10}^{110} \) calculated from eqns. (3) and (4), employing \( a_0 \) equal to 3.142 Å and \( b \) are 913, 383 and 425 eV, respectively; these values of \( E_{10}^{\text{SI}} \) represent upper limits to this quantity and the real values may be smaller by at least a factor of two (see, for example, Thompson 1969) for a "softer" interatomic potential. The corresponding maximum ranges of the RCSs are 86, 55 and 24 Å along the \{111\}, \{110\} and \{100\} directions, respectively; these values were calculated employing Nelson's (1963) expressions for the total number of collisions in a chain. The effect of zero point and thermal motion of the lattice atoms has been shown to decrease the maximum range of RCSs (see, for example, Nelson, Thomson and Montgomery 1962, Sanders and Finit 1964, Tenenbaum 1978). Thus, the above calculated values of the maximum ranges of RCSs would be further decreased by the inclusion of the zero point and thermal motion of the lattice atoms. In the case of our experiments the zero point motion had the dominant effect because of the low irradiation temperatures (10 to 18 K) employed; the Debye temperature of tungsten is 310 K (Lauff 1956).

In a soft atomic collision one has the possibility of generating an RCS (Thompson 1969, Nelson 1968). A simple analytical model of the replacement process suggested that the replacement energy \( E_{10}^{\text{SI}} \) was equal to \( E_{10}^{\text{SI}}/4 \) (Thompson 1969). An atom with an initial energy between \( E_{10}^{\text{SI}} \) and \( E_{10}^{\text{SI}}/4 \) would generate an RCS, whereas one with an energy less than \( E_{10}^{\text{SI}}/4 \) would produce an FC. The value of \( E_{10}^{\text{SI}} \) determines the point in the RCS chain where the SIA is deposited and therefore the length of the chain. This one would expect the range of an RCS to be shorter than the range of an FC. From all the above it is clear that our experimental values of RCSs in tungsten, could be greater by almost a factor of ten than the calculated values of the RCS range. This discrepancy may be a result of inadequacies in the theoretical models of RCSs in a non-static lattice or, alternatively, our sampling procedure as discussed earlier in this section.

5.3. The contribution of focused or replacement collision sequences to sputtering

Current and Seidman (1980) and Current and Seidman (1981) recently completed an extensive study of the sputtering of tungsten surfaces at 10 K, employing the FIM technique. In this research they found that the number of vacancies observed in the near-surface region of the sputtered specimen is approximately consistent with the continuum model of Sigmund (1969 a, b, 1961) for the average theoretical sputtering yield \( \langle N_{\text{vac}}(\theta) \rangle \); the quantity \( N_{\text{vac}}(\theta) \) represents the average number of atoms sputtered per projectile ion at an angle \( \theta \) with respect to the normal to the surface being sputtered.

The result that the mean range of RCSs, in tungsten, may be as large as 20 to 50 Å implies that the mean range of RCSs can have a comparable value. The intersection of an RCS with the surface of a specimen may cause a sputtered atom; a necessary condition is that the last atom in the RCS must have received an energy equal to the binding energy. An RCS cannot result in a sputtered atom if its total chain length is greater than the distance from its site of creation to the surface of the specimen (see, for example, Thompson 1969, 1979, Nelson 1969). A calculation of the exact contribution to the sputtering yield from DZs that do not have a portion of their volume in the near-surface region depends on a knowledge of the range distributions of both RCSs and DZs. The interesting point about our results on tungsten is that we are able to obtain approximate agreement with the continuum sputtering model and also measure a mean range of RCSs that may be as large as 10 to 20 Å.

ACKNOWLEDGMENTS

We thank Mr. R. Whitmarsh for continuing technical assistance and Dr. R. M. Hsue for a careful reading of the manuscript. This work was supported by the United States Department of Energy. Additional support was received from the National Science Foundation through the use of the technical facilities of the Materials Science Center at Cornell University.

APPENDIX

Section for the determination of the positions of the SIAs and depleted zones in an FIM specimen

In this Appendix we describe the details of the procedure for determining the positions of the SIAs with respect to DZs in an FIM specimen. The end-form of the FIM specimen is approximated by a hemisphere of initial radius \( r_0 \) (see fig. 10). As a specimen is successively evaporated atom by atom, a final end-form with an average radius \( r_1 \) is obtained. The centres of the initial and the final hemispheres are denoted by the points O and C respectively. From the FIM micrographs we know the \( \langle k\ell \rangle \) plane and the number of the layer \( n \), in which a given SIA is detected; the value of \( n \) is measured from the initial surface. For example, an SIA at C is found in the nth layer of the \( \langle k\ell \rangle \) plane measured from the initial surface at B. The problem is to determine the coordinates of point C given the quantities \( h_n, k_n, l_n \) and \( n \).
The vector \( \mathbf{v} = [h_2 k_2 l_2] \) is determined from the cross-product
\[
[h_2 k_2 l_2] = [h_3 k_3 l_3] \times [h_1 k_1 l_1].
\]

The \( h, k \) and \( l \) axes and the origin \( O \) therefore constitute a rectangular coordinate system. The coordinates of point \( C \) are given by \((u, v, w)\) in this rectangular coordinate system. The spherical coordinates \((\rho, \theta, \phi)\) of point \( C \) are shown in Fig. 10.

Initially, the centre of the \((h_4 k_4 l_4)\) plane is at \( B \). As a result of the specimen being pulse-field evaporated, the final location of the \((h_4 k_4 l_4)\) plane is at point \( D \). In general, the vector \( \mathbf{BD} \) is tilted away from the \( z\) axis by an angle \( \Delta \theta \). The angle \( \Delta \theta \) was determined by analyzing the triangle \( BDF \); point \( F \) is defined by the expression \( \mid \mathbf{OF} \mid = r_0 \). The length of the vector \( \mathbf{BD} \) is estimated by counting the number of layers (\( n \)) of the \((h_4 k_4 l_4)\) plane which are field-evaporated as the specimen reaches its final end-form:
\[
\rho = |\mathbf{BD}| = |\mathbf{BD}| = r_0. \tag{A 12}
\]

The quantity \( \theta_0 \), the angle between the normal vector \([h_4 k_4 l_4]\) of the \((h_4 k_4 l_4)\) plane and the \( w \) axis, is given by
\[
\theta_0 = \cos^{-1} \left( \frac{h_3 k_3 l_3}{\sqrt{(h^2 + k^2 + l^2)}} \right). \tag{A 13}
\]

The plane \((h_4 k_4 l_4)\) is at \( D \). In general, the vector \( \mathbf{BD} \) is tilted away from the \( z \) axis by an angle \( \Delta \theta \). The angle \( \Delta \theta \) was determined by analyzing the triangle \( BDF \); point \( F \) is defined by the expression \( |\mathbf{OF}| = r_0 \).

The projection of point \( B \) onto the \( w-z \) plane is denoted by point \( P \). The vector \([h_1 k_1 l_1] = \mathbf{OP} \) is therefore given by
\[
|\mathbf{OP}| = \mathbf{BD} + |\mathbf{BF}|. \tag{A 15}
\]
or

\[ \mathbf{b}_1 \times \mathbf{b}_2 = \frac{r_0}{\sqrt{h_1^2 + k_1^2 + l_1^2}} \left( -h_2 k_1 l_2 + k_2 l_1 h_1 + l_2 h_1 k_1 \right). \]

Since the projection of the vector \( \mathbf{b}_1 \times \mathbf{b}_2 \) onto the \( u-v \) plane is parallel to the vector \( (h_2, k_2, l_2) \), we obtain

\[ \phi = \cos^{-1} \left( \frac{h_2 k_1 + k_2 l_1 + l_2 h_1}{\sqrt{(h_1^2 + k_1^2 + l_1^2)(h_2^2 + k_2^2 + l_2^2)}} \right). \]

The spherical coordinates \( (\rho, \theta, \phi) \) of point \( C \) determined by eqs. (A 12), (A 14) and (A 17) are transformed to the rectangular coordinates \( (x, y, z) \) according to the relations

\[ u = \rho \sin \theta \cos \phi \]
\[ v = \rho \sin \theta \sin \phi \]
\[ w = \rho \cos \theta. \]

The rectangular coordinates \( (x, y, z) \) are finally transformed to the standard cubic coordinates \( (x', y', z') \). Thus, from eqns. (A 1) to (A 20) we can calculate the rectangular coordinates \( (x', y', z') \) for any SIA by knowing the values of the quantities \( r_0, r_1, h_1, k_1, l_1, h_2, k_2, l_2, h_3, k_3, l_3, m \) and with these values are obtained from the analysis of EFM micrographs. Above computations are carried out with the aid of our NOVA 1200 computer. The program for these computations is denoted DZP4 (1978) and is available upon request.

References
