Available online at www.sciencedirect.com

SCIENCE DIRECT®

Journal of Nuclear Materials 328 (2004) 77–87

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Review: Evolution of stacking fault tetrahedra and its role in defect accumulation under cascade damage conditions ☆

B.N. Singh [a,*], S.I. Golubov [b], H. Trinkaus [c], D.J. Edwards [d], M. Eldrup [a]

[a] *Department of Materials Research, Risø National Laboratory, DK-4000 Roskilde, Denmark*

[b] *Metals and Ceramics Division, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN-37831-6138, USA*

[c] *Forschungszentrum Jülich, Institut für Festkörperforschung, D-52425 Jülich, Germany*

[d] *Materials Structure and Performance Group, Pacific Northwest National Laboratory, P.O. Box 999, Richland, WA 99352, USA*

Received 10 February 2004; accepted 22 April 2004

Abstract

In order to help understand the evolution of stacking fault tetrahedra (SFTs) in a cascade producing irradiation environment, the available information on the behaviour of SFTs observed under different experimental conditions has been briefly reviewed. Effects of thermal annealing and irradiations on the stability of pre-existing SFTs produced by quenching and aging are also included in the review. Some results on the effects of thermal annealing of irradiation-induced SFTs are presented and discussed. The analysis of these observations leads to three significant conclusions: (a) during irradiation SFTs produced in the cascades are likely to interact with vacancies, self-interstitial atoms (SIAs) and SIA clusters, (b) interaction with SIAs and their clusters may cause both shrinkage and transformation of SFTs into Frank loops and (c) both during irradiation and annealing the lifetime of SFTs is determined not only by their thermal stability but also by their stability against transformation to loops. These facts must be taken into account in the theoretical treatments of damage accumulation.

Published by Elsevier B.V.

Contents

☆ Presented as an invited talk in the Kiritani Memorial Session at the ICFRM-11 held at Kyoto, Dec. 7–12, 2003.

* Corresponding author. Tel.: +45-46 775 709; fax: +45-46 775 758.
E-mail address: bachu.singh@risoe.dk (B.N. Singh).

0022-3115/$ - see front matter Published by Elsevier B.V.
doi:10.1016/j.jnucmat.2004.05.001

1. Introduction

The generation and accumulation of lattice defects and their clusters in various forms have been studied both experimentally and theoretically for more than half a century. The sustained interest and activities in this field have been driven not only by the academic interest in establishing a proper understanding of intrinsic properties of these defects and their agglomerates but also by the practical concern that both physical and mechanical properties of metals and alloys are significantly altered by the presence of these defects and their clusters. The phenomenon of radiation-induced increase in the yield strength (commonly known as radiation hardening) and a severe reduction in work hardening ability and ductility (commonly referred to as low temperature embrittlement) at temperatures below the recovery stage V are well known. These phenomena are, in fact, very good examples of the so-called 'Wigner disease' [1] caused by energetic neutrons. Since the early investigations of radiation hardening already in the 1950s and 1960s [2–5] a large amount of both experimental and theoretical results have become available on this topic (see Ref. [6] for a critical review). Unfortunately, our understanding of these phenomena still remains rather nebulous and a considerable amount of further effort seems to be necessary to elucidate the fundamental issues involved [7].

Another significant effect of neutron irradiation is the formation and growth of voids causing volumetric expansion of materials exposed to strong irradiation environments. The phenomenon of void swelling was discovered already in 1967 [8]. Since then a large number of experiments have demonstrated that both fcc and bcc metals exposed to a strong irradiation environment exhibit this phenomenon [9,10]. In spite of a substantial effort devoted to this topic, various aspects of this problem of void swelling are still not clearly understood.

In recent years a theoretical framework called 'production bias model' (PMB) has been developed to treat the global evolution of damage accumulation including void swelling during cascade producing irradiation. The PBM is based on the concept of production bias, (in addition to dislocation bias) which has its origin in the asymmetric clustering of self-interstitials (SIAs) and vacancies directly in displacement cascades [11]. The PBM in its current form takes into account the consequences of 1-D diffusion of SIA clusters produced in the cascades on changes in reaction kinetics and considers the impact of changes in the direction of motion of SIA clusters while performing 1-D diffusion. Details of these investigations have been published in a number of papers and have been recently summarized in Refs. [12–14]. Very recently, the impact of 2-D diffusion of SIA clusters/loops by conservative climb on defect reaction kinetics has been incorporated in the consideration of 1-D to 3-D diffusion reaction kinetics [15], making the PBM fully capable of describing the damage accumulation under cascade damage conditions.

Even though the specific issues such as the effect of irradiation dose level [16], of recoil energy [17] and of grain size [18] on microstructural evolution including void swelling have been adequately described by the PBM, these calculations did not consider the details of either the evolution or the reaction kinetics of stacking fault tetrahedra (SFTs) during cascade irradiation, simply because it was not absolutely necessary in these first modelling attempts. In a recent attempt to calculate the temperature dependence of void swelling, it became apparent, however, that a number of questions regarding the intrinsic energetic and kinetic properties and the dynamic behaviour of SFTs in the neutron irradiation environment will have to be clarified before the magnitude of damage accumulation can be calculated quantitatively. It was therefore deemed reasonable to consider the basic issues such as nucleation, growth, stability, transformation, shrinkage and annihilation of SFTs in a dynamic irradiation environment. It should be emphasized that this exercise also offers us the possibility of acknowledging and paying tribute to the lifelong contributions of the late Professor Michio Kiritani to this field of research (see later for the specific references).

In Section 2, we first consider the origin and evolution of SFTs which is then followed by the consideration of the evolution of SFTs during irradiation (Section 3). The formation of SFTs during high-speed deformation is briefly described in Section 4. The shrinkage and annihilation behaviour of SFTs are discussed in Section 5. Results of post-irradiation annealing of SFTs in copper are presented in Section 6 and are compared with the annealing results of SFTs in gold produced by quenching. Section 7 discusses various aspects of the evolution of SFT microstructure under dynamic irradiation conditions. A brief summary and some preliminary conclusions of the present work are given in Section 8.

2. Evolution of SFTs during ageing after quenching

One of the most interesting and intriguing microstructural features resulting from the agglomeration of vacancies are the well known stacking fault tetrahedra (SFTs). The formation of SFTs in quenched and aged gold was first discovered by Hirsch and Silcox [19] using transmission electron microscopy (see Ref. [20] for experimental details). The main driving force for the formation of SFTs in quenched and aged metals and alloys is the vacancy supersaturation which is created by first heating the metal close to the melting temperature and then quenching it rapidly down to the vacancy migration temperature. The excess of these non-equilibrium vacancies, will upon aging, at temperatures above the vacancy migration temperature unavoidably agglomerate and form clusters of vacancies. Already in 1950, it was suggest by Frank [21] that the vacancy agglomerates in the form of discs may collapse and form dislocation loops under the condition of vacancy supersaturation. A detailed discussion of the formation and properties of such loops has been reported by Kuhlmann–Wilsdorf [22]. According to Silcox and Hirsch [20], the formation of SFTs occurs by the dissociation of such Frank sessile dislocation loops bounded by a Frank partial dislocation. In an fcc metal of low stacking fault energy the Frank sessile dislocation, having a relatively large Burgers vector, will collapse into a low energy stair–rod dislocation [23,24] and Shockley partial on an intersecting slip plane.

Later, Czjzek et al. [25] proposed another nucleation mechanism according to which SFTs grow by the absorption of single vacancies from a small nucleus containing as few as six vacancies. A similar conclusion has been reached by Kimura et al. [26] and Shirai et al. [27]. De Jong and Koehler [28] suggested that this continuous growth proceeds by the migration of 'ledges' over the faces of the SFTs. The geometry of such ledges has been investigated by Thompson [24] and Seeger and Bross [29]. Both growth and shrinkage of SFTs have been considered in terms of nucleation and motion of ledges by Kimura et al. [26] and Kuhlmann-Wilsdorf et al. [30].

Since the formation of SFTs results from the clustering of vacancies, their density and size would be expected to depend on the level of vacancy supersaturation. Experimental results do indeed confirm that this is the case. Cotterill has shown, for example, that in Au the SFT density increases and size decreases with increasing quenching temperature [31]. A similar variation has been reported for gold with increasing vacancy concentration by Chik and Seeger [32]. Also in gold, Meshii has investigated the effect of ageing temperature both on density and size of SFTs for quenching temperatures of 1148 and 1273 K [33]. The SFT density was found to decrease and the size to increase with increasing ageing temperature, showing again that the nucleation and growth of SFTs are dependent on vacancy supersaturation.

3. Evolution of SFTs during irradiation

It is well established that during irradiation producing multidisplacement cascades, SFTs are formed directly in the cascades or subcascades in fcc metals such as Au, Ag, Cu and Ni [9,34–38]. In recent years, molecular dynamics (MD) simulations have confirmed the formation of SFT directly in cascades in Cu (see Ref. [39] for a recent review). It should be mentioned that the formation of SFTs has been observed in Cu, Ni and Au even during 2 MeV electron irradiation [40], but only in the region close to the 'top' surface where electrons enter the sample. It is also well known that during neutron irradiation at void swelling temperatures (i.e. close to and above recovery stage V) both SFTs and voids are formed and that the density of SFTs (at a given temperature and dose level) is much higher than that of voids [9,35,41]. Furthermore, while voids grow with increasing displacement dose level, the SFTs do not grow in size [35,42]. It is interesting to note that, on the one hand, there has been an extensive amount of discussion in literature about the evolution of voids and void swelling, but the evolution of SFTs has, for some reason, been more or less completely ignored. In order to initiate a discussion on this overlooked issue, we present in the following experimentally known facts about the evolution of SFTs deduced from the post-irradiation observations as well as from a limited number of in situ dynamic observations [43]. This is a necessary prerequisite for a complete and a quantitative description of the kinetics of void swelling under cascade damage conditions in fcc metals and alloys. In the following we shall consider the effect of recoil energy, displacement dose level and irradiation temperature on the density and size of SFTs. The influence of specimen thickness (prior to irradiation) on the evolution of SFTs will be also briefly described.

3.1. Effect of recoil energy

It should be pointed out that only a very limited amount of experimental results is available on this topic. In fact, no systematic studies have been reported from which the effect of recoil energy on SFT evolution can be ascertained for a given temperature, damage rate and displacement dose level. However, in the cases of irradiations of bulk specimens, the available results do not show any significant effect of recoil energy on the density or size of SFTs [35,36,44,45]. The size distributions for the Au and Cu irradiated at temperatures in the range of 300–363 K with fission and fusion neutrons and 600 MeV protons are shown in Fig. 1 [45]. Results for similar irradiation conditions are also reported for Au in Ref. [45]. Similar results have been reported for the bulk specimens of Cu irradiated at 473 and 573 K and Au at 573 K. Clearly, the variation in the recoil energy does not have any significant effect on the size distribution or the average size of the SFTs observed. It should be mentioned, however, that Kiritani et al. [35] have shown that in the case of thin foil irradiations of Au and Ni at $\approx$573 K, there are noticeable differences in the size distributions and the density of SFTs between fission and fusion neutron irradiations.

3.2. Effect of displacement dose level

During neutron irradiation at temperatures both below and above the recovery stage V, the density of SFTs in fcc metals increases with increasing dose level and comes to saturate at some relatively low dose level ($\approx$0.1 dpa in Cu at $\approx$300–573 K) [9,35,38]. A general consensus appears to be that this saturation occurs due to direct impingement of a new cascade on an existing SFT. As a result, the vacancies contained in the SFT are incorporated in the newly generated displacements in the new cascade volume and finally a new SFT emerges at the end of the cooling phase of this new cascade. Recent MD simulations [46] have shown that the direct impingement event simply replaces the old SFT by a new one without production of any significant number of new defects. In other words, beyond the saturation dose level, the effective production rates of SIAs and vacancies are substantially reduced. This would have serious impacts on the dose dependence of the accumulation of both SIAs and vacancies and has to be taken into account in theoretical calculations.

In spite of the fact that the density of SFTs increases very significantly with increasing dose (below the saturation dose) level, the mean size and the size distribution of SFTs do not change to any noticeable extent. Fig. 2 shows size distributions measured in fission neutron irradiated Cu at 373 K to different doses in the range of 0.01–0.3 dpa [47]. The results show that even though there is some indication of a limited amount of growth of SFTs, the effect of irradiation dose is rather insignificant. It is interesting to note that no significant number of SFTs grow beyond $\simeq$6 nm in size at any dose level. Similar results have been reported for Cu irradiated with 14 MeV neutrons at 423 K in the neutron fluence range of 3.1×10^{19}–10^{21} n/m^2 (Fig. 12 in Ref. [36]) and with fission neutrons at 523, 573 and 623 K in the fluence range of $\approx1\times10^{20}$ to $\approx2\times10^{22}$ n/m^2 ($E>$ 1 MeV) [48].

3.3. Effect of irradiation temperature

The experimental results on the temperature dependence of SFT density and size for neutron-irradiated Au, Ag and Ni are very limited. On neutron-irradiated copper, on the other hand, the temperature dependence has been studied systematically and therefore in the following we shall concentrate on the results reported for pure copper.

In fission-neutron-irradiated copper the density of SFT has been found to be strongly temperature

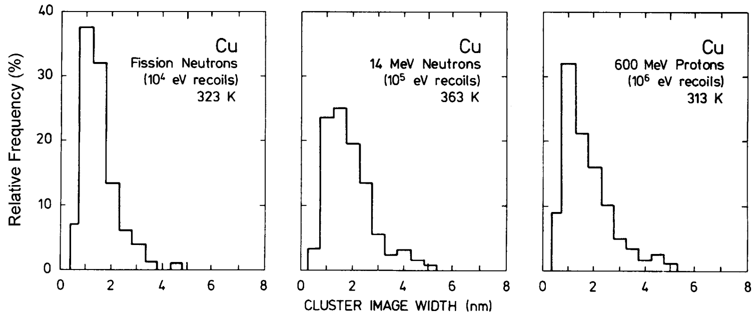

Fig. 1. Size distributions of SFTs in pure copper irradiated at temperatures in the range of 313–363 K with fission neutrons, 14 MeV neutrons and 600 MeV protons (see Ref. [45] for details). Note that the size distribution does not change in any significant way even when the recoil energy is varied by two orders of magnitude. Similar results for gold are shown in [45].

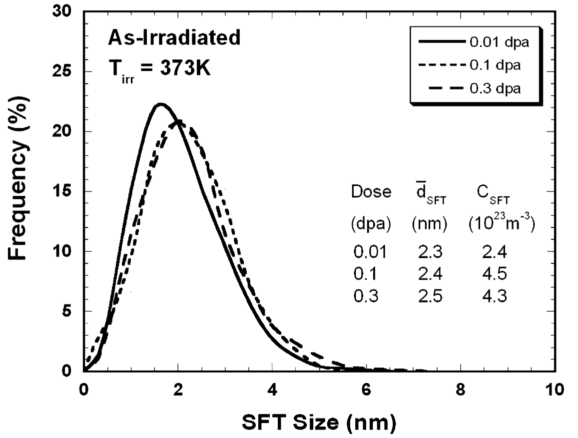

Fig. 2. Size distribution of SFTs (with a bin size of 0.5 nm) in pure copper irradiated with fission neutrons at 373 K to different displacement dose levels in the range of 0.01–0.3 dpa [47]. No significant change in the size distribution is observed even when the dose level is changed by a factor of 30.

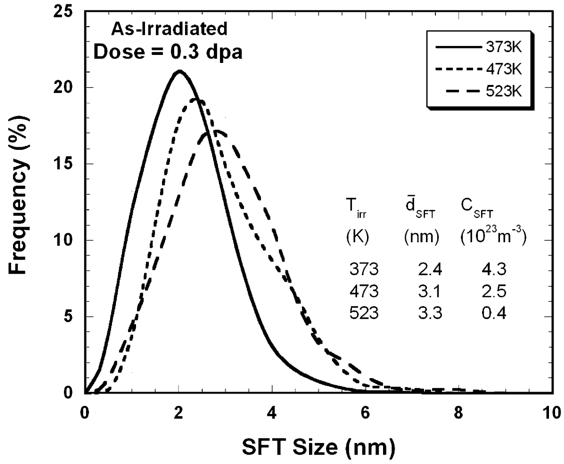

Fig. 3. Effect of irradiation temperatures on size distribution of SFTs (with a bin size of 0.5 nm) in fission neutron-irradiated copper to a dose level of 0.3 dpa. There is a slight change in the peak as well as average sizes with increasing irradiation temperature. Note shrinkage of small ones and growth of large ones with increasing irradiation temperature. However, SFTs do not seem to grow in size beyond about 6 nm.

dependent particularly at temperatures above the recovery stage V [9,41,48]. A similar temperature dependence has been observed in copper irradiated with 14 MeV neutrons [35] at temperatures between 300 and 563 K. Although the results shown in Ref. [9] yield an 'effective' activation energy of 0.85 eV, the physical process and the mechanism controlling this temperature dependence still remains unclear and unidentified. Recent results on the temperature dependence of SFT density obtained from post-irradiation annealing experiments indicate an apparent activation energy of ≈1.6 eV [49]. The interpretation of this energy also remains uncertain.

Fig. 3 shows the temperature dependence of size distributions of SFTs for pure copper irradiated with fission neutrons to a dose level of 0.3 dpa at 373, 473 and 523 K. Once again neither the shape of the size distribution nor the average size of SFTs are affected to any significant extent by the irradiation temperature. As mentioned above, the density of SFTs is, on the other hand, strongly affected by irradiation temperature. Note that even at 523 K not many SFTs are found to grow beyond 6 nm in size. Kiritani et al. [35] have also shown that the size distributions of SFTs in copper irradiated with fission and fusion neutrons remain practically the same at 473 and 573 K. Similar results have been reported for neutron irradiated copper by English et al. [48].

3.4. Evolution of SFTs in thin foil and bulk samples

From the point of view of establishing a proper understanding of the evolution of SFTs under dynamic conditions of cascade producing irradiation, it is interesting to consider the results reported for thin foil and bulk samples [35,44] of pure metals irradiated with fission and fusion neutrons. The basic idea behind these valuable experiments was to examine the impact of SIAs and their clusters produced in cascades on the evolution of SFTs. These experiments have established two important facts. First, the density of SFTs in Cu, Au and Ni is considerably higher (at a given dose level and irradiation temperature) in the thin foil than that in the bulk sample (see Figs. 30–32 in Ref. [35] and Fig. 12 in Ref. [44]). Secondly, the rate of increase of SFT density with dose is considerably higher in the case of thin foil than that in the bulk sample. This behaviour is found to be the same both under fission and fusion neutron irradiations. It is also interesting to note that the size distribution and the average size of SFTs in Au, irradiated as thin foils, for example, are temperature sensitive. In other words, the size distribution moves towards larger sizes and the average size becomes larger with increasing temperature (see Fig. 22 in Ref. [35]). This annealing behaviour is remarkably different from that observed in the case of bulk Au.

The observed differences in the SFT evolution behaviour between thin foil and bulk irradiations have been considered by Kiritani et al. [35,44,50,51] to arise due to shrinkage of SFTs caused by their interactions with freely migrating SIAs and their clusters. In the case of thin foil irradiations, most of the SIAs and their clusters escape to specimen surfaces (i.e. without affecting SFTs) whereas in the bulk irradiations they interact with the high density of SFTs present in the bulk. These interactions lead to the shrinkage of SFTs in the bulk.

A similar conclusion has been drawn from the results of in situ heavy-ion irradiation experiments [43].

4. Production of SFTs during high-speed deformation

While discussing the evolution of SFTs by condensation of vacancies, it is worth considering the formation of SFTs during high-speed deformation. This is of a particular relevance since this was the last important and exciting discovery made by the late Professor Michio Kiritani. The historical background and details of the techniques of deformation of thin films to fracture and high-speed deformation of bulk samples have been summarized by Kiritani himself [52,53]. A large amount of work carried out in this area has been published as a complete volume of Materials Science and Engineering in 2003 (see Ref. [53]).

The main relevance of the work on high-speed deformation to the present topic is the formation of a high density of SFTs in Au, Cu, Ni and even in Al [54] in which SFTs have never been observed neither by irradiation nor by quenching and aging. These results emphasize the importance of vacancy supersaturation for the formation and growth of SFTs. Kiritani et al. [54] have argued that the high vacancy supersaturation during high-speed deformation arises due to the process of parallel shifts of atomic planes and not by dislocation generation. MD simulations have demonstrated, on the other hand, that a very high concentration of vacancies is likely to be created by the generation and interactions of dislocations during high-speed deformation [55].

5. Shrinkage and annihilation of SFTs

5.1. Thermal annealing of 'fully grown' SFTs

It has been shown experimentally that 'fully grown' SFTs are thermally stable, for example, in Au up to about 873 K [31,56,57]. By determining the size of the smallest tetrahedra (surviving at a given annealing temperature) as a function of annealing temperature, Clarebrough et al. [58] have shown that the smaller SFTs in gold anneal out at somewhat lower temperature ($\simeq$823 K).

As regards the question of operating mechanism during annealing, no single mechanism has been identified either experimentally or theoretically. Kimura et al. [26] and Kuhlmann-Wilsdorf et al. [30] have suggested that a 'fully grown' SFT could not be dissolved by successive thermal emission of vacancies. Kuhlmann-Wilsdorf has considered the possibility of shrinkage by the migration of interstitial ledges nucleated at corners of SFTs [59]. On the basis of an estimate of vacancy undersaturation required for the shrinkage to take place, it was concluded that this was unlikely to be the operating mechanism. A two-step mechanism involving first growth of SFTs to a metastable size by the migration of vacancy ledges nucleated at corners and then collapse of the SFT into prismatic loops which are then eliminated by climb has been also considered by Kuhlmann-Wilsdorf [59]. Meshii and Kauffman [56] have assumed that a Shockley partial dislocation loop nucleate on one of the faulted planes of a SFT and grows until it causes the collapse of the SFT into a Frank sessile loop which finally dissolves by emitting vacancies.

On the basis of experimental observations also Kiritani [40] has concluded that the SFTs in gold do not shrink by vacancy emission instead they first convert into faulted loops and then shrink by vacancy emission. On the basis of TEM results on annealing of SFTs in gold, Washburn [60] has also concluded that 'SFTs do not shrink as tetrahedra but *always* collapse into triangular Frank loops'. Shrinkage then occurs by emission of vacancies from the loops.

5.2. Shrinkage of SFTs by irradiation-induced SIAs

It is very interesting to note that it was already in 1961 that Silcox [61] had proposed that SFTs could collapse by absorbing a sufficient number of SIAs. In order to test this hypothesis, Hirsch et al. [62] carried out irradiation experiments on $\simeq$100 nm thick TEM specimens of Au containing 1.4×10^{21} SFT/m^{-3} (produced by quenching and aging). Irradiations were carried out with 1.5 and 3.5 MeV α-particles at 293 K (i.e. considerably below the recovery stage V, at about 575 K) to different dose levels. Their TEM results did indeed confirm that the SFTs annihilated as a result of irradiation. Interestingly enough they found that in fact SFTs do not anneal out as SFTs but collapse into loops, causing an increase in the density of so-called 'black spot defects' (see Fig. 2 in Ref. [62]).

Following these experiments by Hirsch et al. [62], Howe and McGurn [63] conducted more irradiation experiments to obtain further information on the mechanism of collapse of the SFTs formed during quenching and annealing. They irradiated Au foils containing SFTs produced by quenching from 1223 K into brine at 273 K and aging for 1 h at 373 K. Irradiation was carried out at room temperature with 100 keV O^- ions which led to disappearance of the SFTs. Similar disappearance (collapse) of SFTs was observed during bombardment of Au specimens containing SFTs with 100 keV O^-ions at temperatures even below 30 K.

Venables and Balluffi [64] irradiated quenched and aged foils of Au with 200 eV Au^+ ions at 143 and 303 K. Note that the irradiation with 200 eV Au^+ ions produces SIAs $\approx$10 nm below the surface by replacement collision sequences. They found that the irradiation above 283 K (i.e. where SIAs are mobile) led to disappearance of SFTs.

Finally, it should be mentioned that Kiritani [40] has irradiated Au foils containing SFTs with 1 MeV electrons at 298 K and found that the SFTs shrink as SFTs (i.e. without transformation to loops) by absorbing SIAs.

6. Post-irradiation annealing of SFTs

Since practically nothing is known (at least to our knowledge) about the thermal annealing behaviour of irradiation produced SFTs in copper, we have carried out a series of isothermal annealing experiments on neutron irradiated copper. Specimens were irradiated in the DR-3 reactor at Risø National Laboratory (see Ref. [47] for details) to different dose levels and at different temperatures. The size distributions measured in the as-irradiated condition are presented already in Figs. 2 and 3. In the following we present the results of post-irradiation annealing experiments on (a) specimens irradiated at 373 K to different displacement dose levels which were annealed at 573 K for 50 h (Fig. 4) and (b) specimens irradiated to a dose level of 0.3 dpa at 473 K and then annealed at different temperatures (Fig. 5).

Results shown in Fig. 4 demonstrate that the post-irradiation annealing behaviour of SFT is rather complicated. The annealing behaviour of SFTs produced at 0.01 dpa, for instance, is clearly different from those produced at 0.3 dpa. Even though in both cases a very large fraction of small (<2 nm) SFTs anneal out during annealing, the SFTs larger than ≈3.5 nm produced at 0.3 dpa show some limited amount of growth and the average SFT size increases from 2.4 nm (as-irradiated) to 4.0 nm after annealing at 573 K for 50 h. In neither of the two cases, however, did any significant number of SFTs grow beyond ≃6 nm in size. Clearly, the annealing behaviour is not being controlled by a simple process such as classical Ostwald ripening, even though the number of vacancies accumulated in SFTs produced at 0.1, 0.2 and 0.3 dpa are conserved during annealing [47] which is one essential feature of Ostwald ripening. It should be noted that the annealing leads to a limited increase both in the peak size as well as the average SFT size in specimens irradiated to 0.01 as well as 0.3 dpa. It is also worth pointing out that the density of SFT is significantly reduced by annealing of specimen irradiated to 0.01 dpa but a relatively smaller decrease occurred during annealing of the specimen irradiated to 0.3 dpa.

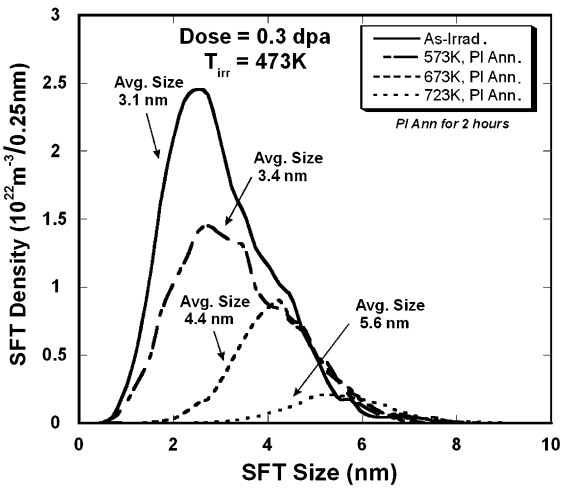

Fig. 5. Effect of post-irradiation annealing temperature on the size distribution of SFT in copper irradiated at 473 K to a dose level of 0.3 dpa; annealing was carried out for 2 h at all three temperatures. For comparison, the size distribution for the as-irradiated copper is also shown. Both the peak size and the average size increase with increasing annealing temperature. It is interesting to note that in none of the cases SFTs grow beyond the 6–8 nm size range.

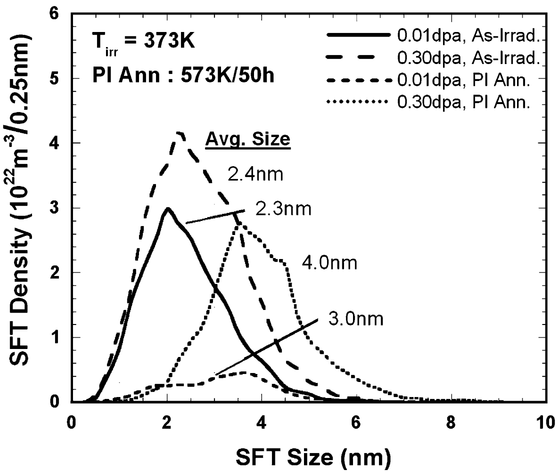

Fig. 4. Effect of post-irradiation annealing (at 573 K for 50 h) on the size distribution of SFTs in pure copper irradiated at 373 K to different dose levels. Note that the annealing behaviour of specimens irradiated to 0.3 dpa is significantly different from the specimen irradiated at 0.01 dpa. In the former case, the small SFTs shrink and the larger ones grow but not beyond ≃7 nm. It should be pointed out that in the case of 0.01 dpa specimen, annealing causes a loss of vacancies whereas in the 0.3 dpa specimen, the number of vacancies is conserved.

Fig. 5 shows SFT size distributions measured in specimens irradiated at 473 K to 0.3 dpa and then annealed for 2 h at 573, 673 and 723 K. The size distribution for the SFTs in the as-irradiated condition is also shown. The density of SFTs decreases and both the peak size and the average size of SFTs increase with increasing annealing temperature. At 723 K, practically all SFT's up to 3 nm in size completely anneal out. However, this annealing of smaller SFTs does not lead to any significant growth of the larger SFTs. The fraction of

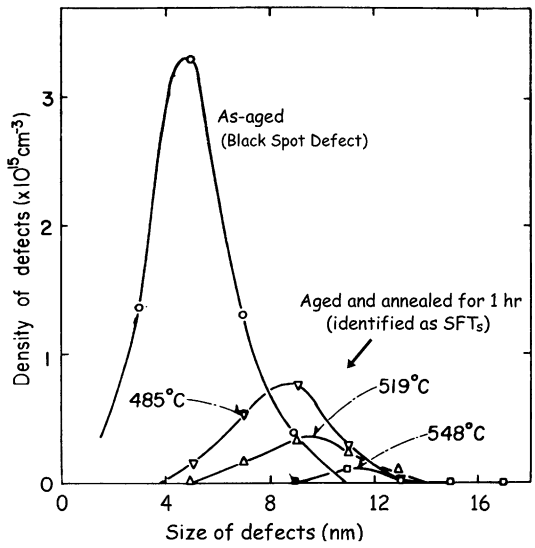

Fig. 6. Size distribution of SFTs in pure gold as a function of annealing temperature [57]. Specimens were quenched and aged prior to annealing. Annealing was carried out at given temperatures for 1 h. General features of the annealing behaviour is very similar to that in Fig. 5. As in the case of copper (Fig. 5) the SFTs do not grow beyond a certain size.

larger SFTs (e.g. beyond 6 nm in size) remains very small for all annealing temperatures. This behaviour again suggests that the annealing of these SFTs is not being controlled by a simple mechanism such as Ostwald ripening.

It is interesting at this juncture to compare the present results of annealing of SFTs in Cu with the results of annealing of SFTs in Au produced by quenching and ageing [57] shown in Fig. 6. The size distribution marked 'As aged' represents the size distribution of the so-called black spot defect clusters after ageing at 373 K. These clusters are some kind of vacancy clusters/loops which then transform into SFTs during annealing at temperatures between 423 and 473 K (see Fig. 5 in Ref. [57]). The annealing at 485 °C (758 K) and above represents the annealing behaviour of SFTs. The similarity between the annealing behaviour of SFTs in Au produced by quenching and ageing (Fig. 6) and that of the SFTs produced in Cu by irradiation is quite obvious. In both cases, the smaller SFTs shrink and anneal out but larger SFTs do not grow beyond a certain size, indicating once again that the annealing mechanism is more complicated than the Ostwald ripening.

7. Discussion

The results reviewed in the preceding sections reveal the degree of complexity involved in dealing with the problem of evolution of SFTs. In order to help the discussion of implications of these complications to the consideration of damage accumulation during a dynamic irradiation experiment, we have summarized the available information regarding the salient features of nucleation, growth, shrinkage and annihilation of SFTs under various conditions schematically in Fig. 7. It should be emphasized that all the information given in Fig. 7 is based on experiments, computer simulations and/or theoretical analysis, except for the case of dynamic irradiation experiment (i.e. the case of 'continued cascade irradiation'). In this box, we mention those events and processes that *can be expected* to occur on the basis of the available information (as reviewed in the previous sections).

Let us now consider the case of a real dynamic neutron irradiation experiment on pure copper, where single vacancies, single SIAs, glissile and sessile SIA clusters and SFTs are being produced continuously and concurrently. Immediately after their production, the SFTs become active sinks for single vacancies, single SIAs and gliding SIA clusters.

Exactly how a newly generated SFT in a cascade may interact with vacancies, SIAs and SIA clusters is likely to depend on its form. A completely perfect SFT would be less effective as a sink than one which is truncated or contains a ledge of vacancy or interstitial type. It is worth mentioning here that Schäublin et al. [65] have recently investigated the form of SFTs in 600 MeV proton irradiated Cu by the combination of TEM and image simulation using MD. They claim to have found about 36% of SFTs produced by irradiation to be truncated. Statistically, it seems rather unlikely that at the end of the cooling down phase every cascade will have exactly the magic number of vacancies required to form a perfect SFT of a certain size. Recently, Bacon et al. [66] and Voskoboinikov et al. [67] have analysed a large number of SFTs formed during MD simulations of 25 keV cascades in Cu and have concluded that most of the SFT-like structures are not regular but are either truncated or consist of more than one SFT joined along a face. In other words, SFT-like structures do not contain exact number of vacancies to form a regular SFT. Anyhow, as discussed in Section 5.2, the freshly produced SFTs may grow by absorbing vacancies and may shrink by absorbing SIAs. Furthermore, the accumulation of SIAs on SFT faces, either by single SIAs or by their clusters, may cause transformation of SFTs into vacancy loops (see Section 5.2). The conclusion that SIA clusters interact with SFTs and destroy them is fully consistent with the results of the in situ, heavy-ion experiments [43]. The same conclusion has been reached by the analysis of thin foil and bulk irradiation experiments (see Section 3.4). Finally, recent MD simulations have demonstrated that the direct impingement of a cascade on a pre-existing SFT leads to dissolution of the

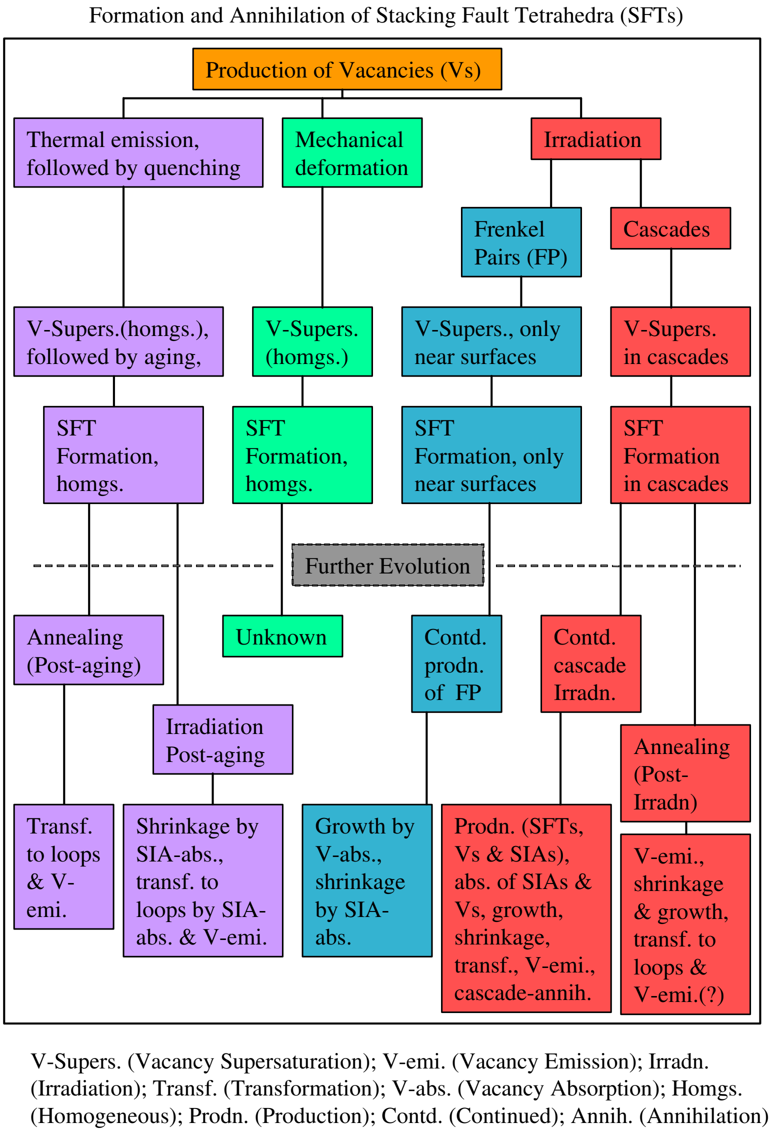

Fig. 7. Schematic illustration of conditions under which SFTs are formed. It shows, first of all, that vacancy supersaturation is the necessary condition for the formation and growth of SFTs. Different processes involved in further evolution of SFTs during annealing (post-quenching and aging and post-irradiation) are identified. It should be noted that both thermal annealing and irradiation can cause transformation of SFTs into Frank loops.

old SFT and formation of a new one [68]. Naturally, the frequency of the impingement event will increase with increasing density of SFTs (for a given production rate of cascades) reaching the maximum when the spacing between SFTs becomes the same as the cascade size.

The scenario outlined above regarding interactions between SFTs and SIAs and their clusters and the ensuing consequences, leads to a fundamentally important conclusion that the lifetime of SFTs produced in cascades is determined not only by their intrinsic thermal stability but also by their interaction with SIAs and the gliding clusters of SIAs. This implies that during an irradiation experiment, the population of SFTs remains really in a dynamic quasi-steady state: they form, grow, shrink, transform and shrink and get annihilated by cascades. In other words, a post-irradiation TEM picture of SFTs represents only a snap-shot of the SFTs population existing at the end of the irradiation experiment. This may explain as to why the size distribution of SFTs does not change in any significant way as a function of recoil energy, displacement dose level and irradiation temperature (Figs. 1–3).

Both irradiation and post-irradiation annealing experiments indicate (Figs. 1–3 and 4,5, respectively) that SFTs are unable to grow in size beyond $\simeq$(6–8) nm. At present no definitive explanation can be given as to why and how this threshold size range is maintained. It could be, however, that the sum of line energy and stacking fault energy makes a growing SFT energetically less favourable than a loop when it reaches a certain size. Kuhlmann-Wilsdorf [59] has considered the possibility of growth by migration of vacancy ledges at corners and subsequent collapse of SFTs. This would suggest that 6–8 nm is the critical SFT size range in copper. When the SFTs reach this size, they collapse into loops and shrink by vacancy emission.

8. Summary and conclusions

The available information on the evolution of SFTs under various conditions such as quenching and ageing, high-speed deformation, electron irradiation and cascade irradiation has been briefly reviewed. In all four cases a certain level of vacancy supersaturation is found to be the necessary condition for the formation and growth of SFTs. It is still not completely certain whether the nucleation of SFTs occurs by first condensation of vacancies into loops followed by their transformation into SFTs or by a simple agglomeration of vacancies.

It has been shown that SFTs can grow by absorption of single vacancies and shrink by absorption of single SIAs.

Accumulation of SIAs on SFT surfaces may lead to the transformation of SFTs into Frank loops. Irradiations of pre-existing SFTs (formed initially by quenching and ageing) have been found to annihilate SFTs by transformation. In situ heavy ion irradiation and irradiation of thin foils have shown that SIA clusters produced in cascade annihilate SFTs.

Even during thermal annealing, 'fully grown' SFTs have been found to shrink not as SFTs by vacancy emission but by collapse into loops which possibly anneal out by vacancy evaporation. The conditions for the transformation of SFTs to loops are not clear. During neutron irradiation the SFT size distribution does not change significantly as a function of recoil energy, displacement dose and irradiation temperature.

Post-irradiation annealing experiments have shown that the small SFTs anneal out but larger ones do not seem to grow beyond a certain limit (in Cu $\simeq$6–8 nm). In other words, shrinkage and growth of SFTs seem not to follow a simple Ostwald ripening mechanism.

The analysis of experimental and MD simulation results clearly suggest that the evolution of SFTs during irradiation with cascade producing particles is likely to be a very complex process involving formation, growth, shrinkage and annihilation of SFTs. The interactions with single vacancies, SIAs and gliding SIA clusters are likely to play a significant role in the damage accumulation. Consequences of these interactions for the reaction kinetics of SFTs must be carefully considered in the theoretical treatments of damage accumulation.

The traditional concept of the thermal stability of SFTs in terms of dissociation energy for single vacancies from the SFTs does not appear to be appropriate and needs to be revised.

The role of cascade resolution of SFTs by cascade impingement in the evaluation of damage production efficiency and damage accumulation needs a careful assessment.

Acknowledgements

This work was supported by the European Fusion Technology Programme. D.J. Edwards would like to thank Risø National Laboratory for the support and assistance during his visit. His work was partly supported by the US Department of Energy under contract DE-AC06-76RLO 1830 with the Battelle Memorial Institute at the Pacific Northwest National Laboratory.

References

[1] E.P. Wigner, J. Appl. Phys. 17 (1946) 857.
[2] A.W. McReynolds, W. Augustiniak, M. McKewon, D.B. Rosenblatt, Phys. Rev. 98 (1955) 418.
[3] T.H. Blewitt, R.R. Coltman, R.E. Jamison, J. Redman, J. Nucl. Mater. 2 (1960) 277.
[4] M.J. Makin, in: W.F. Sheely (Ed.), Radiation Effects, Metallurgical Society Conference Asheville, NC September 1965, AIME, vol. 37, Gordon and Breach, New York, 1966, p. 627.
[5] J. Diehl, in: Vacancies and Interstitial in Metals, in: A. SeegerD. Schumacher, W. Schilling, J. Diehl (Eds.), Proceedings of International Conference on KFA Jülich, 1968, North Holland, Amsterdam, 1969, p. 739.
[6] B.N. Singh, A.J.E. Foreman, H. Trinkaus, J. Nucl. Mater. 249 (1997) 103.
[7] B.N. Singh, N.M. Ghoniem, H. Trinkaus, J. Nucl. Mater. 307–311 (2002) 159.
[8] J. Cawthorne, E.J. Fulton, Nature 216 (1967) 575.
[9] B.N. Singh, S.J. Zinkle, J. Nucl. Mater. 206 (1993) 212.
[10] B.N. Singh, J.H. Evans, J. Nucl. Mater. 226 (1995) 277.
[11] C.H. Woo, B.N. Singh, Philos. Mag. A65 (1992) 889.
[12] H. Trinkaus, B.N. Singh, S.I. Golubov, J. Nucl. Mater. 283–287 (2000) 89.
[13] B.N. Singh, H. Trinkaus, S.I. Golubov, in: K.H.J. Buschow, R.W. Cahn, M.C. Flemings, M.C. Ilschner, E.J. Kramer, S. Mahajam (Eds.), Encyclopedia of Materials: Science and Technology, vol. 8, Pergamon, Oxford, 2001, p. 7957.
[14] H. Trinkaus, H.L. Heinisch, A.V. Barashev, S.I. Golubov, B.N. Singh, Phys. Rev. B 66 (2002) 060105(R).
[15] H. Trinkaus, B.N. Singh, S.I. Golubov, submitted to Philos. Mag. A.

[16] B.N. Singh, S.I. Golubov, H. Trinkaus, A. Serra, Y.N. Osetsky, A.V. Barashev, J. Nucl. Mater. 251 (1997) 107.
[17] S.I. Golubov, B.N. Singh, H. Trinkaus, Philos. Mag. A 81 (2001) 2533.
[18] B.N. Singh, M. Eldrup, S.J. Zinkle, S.I. Golubov, Philos. Mag. A 82 (2002) 1137.
[19] P.B. Hirsch, J. Silcox, in: R.H. Doremus, B.W. Roberts, David Turnbull (Eds.), Growth and Perfection of Crystals, John Wiley, New York, 1958, p. 262.
[20] J. Silcox, P.B. Hirsch, Philos. Mag. 4 (1959) 72.
[21] F.C. Frank, Symposium Plastic Deformation Cryst. Solids, Carnegie Institute of Technology, Pittsburgh, Pennsylvania, 1950, p. 150.
[22] D. Kuhlmann-Wilsdorf, Philos. Mag. 3 (1958) 125.
[23] M.J. Whelan, PhD dissertation, University of Cambridge, 1958.
[24] N. Thompson, Rept. Conf. Defects Cryst. Solids, Physical Society, London, 1955, p. 153.
[25] G. Czjzek, A. Seeger, S. Mader, Phys. Stat. Solids 2 (1962) 558.
[26] H. Kimura, D. Kuhlmann-Wilsdorf, R. Madin, Appl. Phys. Lett. 3 (1963) 4.
[27] Y. Shirai, K. Furukawa, J. Takamura, W. Yamada, S. Iwata, Appl. Phys. A 37 (1985) 65.
[28] M. de Jong, J.S. Koehler, Phys. Rev. 129 (1963) 40.
[29] A. Seeger, H. Bross, J. Phys. Chem. Solids 16 (1960) 253.
[30] D. Kuhlmann-Wilsdorf, H. Kimura, R. Maddin, J. Appl. Phys. 35 (1964) 2557.
[31] R.M.J. Cotterill, Philos. Mag. 6 (1961) 1351.
[32] K.P. Chik, A. Seeger, in: Proceedings of Third European Regional Conference on Electron Microscopy, vol. 1, Publishing House, Czechoslovak Acad. of Sciences, 1964, p. 235.
[33] M. Meshii, in: R.M.J. Cotterill, M. Doyama, J.J. Jackson, M. Meshii (Eds.), Lattice Defects in Quenched Metals, Academic Press, New York, London, 1965, p. 387.
[34] M. Kiritani, Mater. Sci. Forum 15–18 (1987) 1023.
[35] M. Kiritani, T. Yoshiie, S. Kojima, Y. Satoh, K. Hamada, J. Nucl. Mater. 174 (1990) 327.
[36] M. Kiritani, J. Nucl. Mater. 206 (1993) 156.
[37] M. Kiritani, Radiat. Eff. Def. Solids 148 (1999) 233.
[38] S.J. Zinkle, Radiat. Eff. Def. Solids 148 (1999) 447.
[39] D.J. Bacon, Yu. N. Osetsky, R.E. Stoller, R.E. Voskoboinikov, J. Nucl. Mater. 323 (2003) 152.
[40] M. Kiritani, Point Defects and Defect Interactions in Metals, Yamada Science Foundation, Tokyo University, 1982, p. 59.
[41] B.N. Singh, A. Horsewell, P. Toft, D.J. Edwards, J. Nucl. Mater. 224 (1995) 131.
[42] S.J. Zinkle, B.N. Singh, J. Nucl. Mater. 283–287 (2000) 306.
[43] S. Ishino, J. Nucl. Mater. 206 (1993) 139.
[44] J.M. Kiritani, J. Nucl. Mater. 251 (1997) 237.
[45] A. Horsewell, B.N. Singh, S. Proennecke, W.F. Sommer, H.L. Heinisch, J. Nucl. Mater. 179–181 (1991) 924.
[46] R.E. Voskoboinikov, Y.N. Osetsky, D.J. Bacon, to be published.
[47] B.N. Singh, D.J. Edwards, P. Toft, J. Nucl. Mater. 299 (2001) 205.
[48] C.A. English, B.L. Eyre, J.W. Muncie, Harwell report AERE-R-12188, 1986;
Philos. Mag. A 56 (1987) 453.
[49] D.J. Edwards, M. Eldrup, B.N. Singh, S.I. Golubov, to be submitted to Philos. Mag.
[50] M. Kiritani, J. Nucl. Mater. 137 (1986) 261.
[51] M. Kiritani, T. Yashiie, S. Kojima, J. Nucl. Mater. 141–143 (1986) 626.
[52] M. Kiritani, Radiat. Eff. Def. Solids 148 (1999) 233.
[53] M. Kiritani, Mater. Sci. Eng. A 350 (2003) 1.
[54] M. Kiritani, Y. Satoh, Y. Kizuka, K. Arakawa, Y. Ogasawara, S. Arai, Y. Shimomura, Philos. Mag. Lett. 79 (1999) 797.
[55] J. Schiøtz, T. Leffers, B.N. Singh, Philos. Mag. Lett. 81 (2001) 301.
[56] M. Meshii, J.W. Kauffman, Philos. Mag. 5 (1960) 939.
[57] A. Yoshinaka, Y. Shomomura, M. Kiritani, S. Yoshida, Jpn. J. Appl. Phys. 7 (1968) 709.
[58] L.M. Clarebrough, R.K. Segall, M.H. Loretto, M.E. Hargreaves, Philos. Mag. 9 (1964) 377.
[59] D. Kuhlmann-Wilsdorf, in: R.M.J. Cotterill, M. Doyama, J.J. Jackson, M. Meshii (Eds.), Lattice Defects in Quenched Metals, Academic Press, New York, London, 1965, p. 269.
[60] J. Washburn, in: Radiation-Induced Voids in Metals, J.W. Corbett, L.C. Ianniello (Eds.), Proceedings of the 1971 International Conference, held at Albany, New York, 9–11 June 1971, p. 647.
[61] P. Silcox, PhD thesis, Cambridge University, 1961.
[62] P.B. Hirsch, R.M.J. Cotterill, M.W. Jones, in: Proceedings of 5th International Congress on Electron Microscopy, Academic Press, New York, 1962, p. F-3.
[63] L.M. Howe, J.F. McGurn, Appl. Phys. Lett. 4 (1964) 99.
[64] J.A. Venables, R.W. Balluffi, Bull. Am. Phys. Soc. (II) 9 (1964) 295.
[65] R. Schäublin, N. Baluc, M. Victoria, Philos. Mag., in press.
[66] D.J. Bacon, Y.N. Osetsky, R. Stoller, R.E. Voskoboinikov, J. Nucl. Mater. 323 (2003) 152.
[67] R.E. Voskoboinikov, Y.N. Osetsky, D.J. Bacon, to be submitted to Philos. Mag.
[68] Y.N. Osetsky, R.E. Voskoboinikov, D.J. Bacon, to be published.