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THE INTERACTION OF HVEM-GENERATED POINT-DEFECTS WITH DISSOCIATED DISLOCATIONS

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ABSTRACT

This paper describes experiments we have performed to investigate the mechanisms of interaction of HVEM-generated point-defects with dissociated dislocations in a series of austenitic Fe-Ni-Cr alloys and reviews earlier work in Cu-Al alloys and in Ag. In the Fe-Ni-Cr alloys interstitial climb was observed only at favourable sites such as pre-existing jogs, whilst vacancies clustered near dislocations to form stacking-fault tetrahedra. These observations are similar to those in Ag; the complex climb mechanisms seen in Cu-Al alloys were not found. The differences between materials is believed to be due to differences in the case of interstitial pipe diffusion.

1. INTRODUCTION

The interaction of radiation-produced point defects with dislocations is central to the understanding of phenomena which give rise to dimensional or mechanical-property changes in nuclear reactors. As part of a more general effort to understand microstructural evolution under irradiation, studies of the mechanisms of interaction of HVEM-generated point-defects with dissociated dislocations have been carried out in Oxford, Harwell and Argonne over the past 11 years. Various model materials have been chosen, starting with very low stacking-fault energy (SFE) CuAl alloys with wide partial separations and progressing towards the structural steels of practical importance. These studies have demonstrated that dislocation climb in these materials may be far from straightforward. In this paper we briefly review early studies in CuAl alloys and in Ag and present some of our more recent results in Fe-Ni-Cr alloys, The observations in the different materials will be compared and contrasted and possible origins for differences and similarities between materials will be discussed.

2. EXPERIMENTAL

Most of the experiments described here were conducted as follows. Electrochemically-thinned single-crystal foils with foil normals closely parallel to <111> and containing a moderate density of line dislocations were first examined in a conventional 100kV/200kV transmission electron microscope. Line dislocations lying in the plane of the foil were imaged using the weak-beam technique, employing the {220} diffraction vectors available at this pole. The Burgers vectors $\bar{b}_p$ of the individual partial dislocations were determined by use of the Howie and Whelan $\bar{g}.\bar{b} = 0$ criterion. The specimens were then transferred to the Oxford or Argonne HVEM and were held at controlled temperatures whilst the regions containing the characterised dislocations were irradiated with high-energy electrons (generally 1MeV). The irradiations were usually terminated when definite visible signs of radiation damage were evident, for example, the appearance of dislocation loops or a change in appearance of the line. Typically this took about two minutes with an electron flux of 2 x 10^{23} electrons $m^{-2}s^{-1}$. Some irradiations were however carried out to lower or higher doses. The levels of displacement damage (in displacements per atom, dpa) were calculated using values of the displacement cross sections for Frenkel pair production tabulated by Oen [1]. These values were typically in the range 0.02-0.15dpa. Finally, the specimens were returned to a conventional TEM for characterisation of the irradiation induced microstructures. The irradiated dislocations were imaged by the weak-beam technique using a large number of different diffraction vectors, and a detailed diffraction contrast analysis was performed. Throughout this paper Burgers vectors are described using Thompson tetrahedron notation.

3. COPPER-ALUMINIUM ALLOYS

The climb mechanisms in Cu-13%Al, an alloy with a dissociation width of partial dislocations ranging from 20-40nm. for screw-edge dislocation orientations, were first elucidated by Cherns, Hirsch and Saka [2] in a series of definitive experiments. The observation of the formation of Shockley dipoles along irradiated dislocations was explained by supposing the nucleation of interstitial perfect prismatic loops, with Burgers vectors $\bar{b} = {}^1/_2 <110>$, onto individual partials; the partial of greater edge character being favoured. A striking example of loop nucleation, after low dose irradiation, on individual partials is shown in figure 1.

Figure 1: Loops nucleated directly onto individual partials of a 60° dislocation in Cu-13%Al after low-dose, room-temperature irradiation. Micrograph courtesy D. Cherns.

At higher doses subsequent interaction (described fully in the paper of Cherns et al [2]) of the decorated dislocation with the osmotic climb force, which arises from an expected high super saturation of interstitials at dislocations [3,4], was believed responsible for the creation of extremely complex post-irradiation configurations.

Other detailed contrast experiments have established that climb mechanisms similar to those isolated in Cu-13%Al operate for interstitial climb in Si and GaAs under electron irradiation [5,6]. In a higher SFE Cu-10%Al alloy, the evolution of dislocation microstructures under irradiation [7] also appeared consistent with observations in Cu13%Al. In none of these cases was any vacancy component of the damage identified.

4. SILVER

Whilst the SFE's of silver and Cu10%Al are similar [8], dislocations in the two materials respond differently to irradiation: Under room-temperature electron irradiation, dislocations in silver generally seem to constrict and promote in their vicinity the formation of dislocation loops and (especially) stacking-fault tetrahedra (SFT) [9]. There was no indication of major climb motion nor of complex interactions between the clusters and line dislocations. At first there was some confusion regarding the nature of the SFT, as discussed in reference [9]. Initial experiments using the $2^1/_2$D technique [10] suggested that the nature was interstitial [11]. Several subsequent experiments have now however indicated that they are vacancy. These include the observation that SFT form within the inner peripheries of large interstitial loops produced by prior electron irradiations at elevated temperature, shown in figure 2. Since this is a region of compressional strain the defects are likely to be vacancy. Similar results have been reported by Sigle [12]. Vacancy SFT have also been found in high resolution experiments in electron-irradiated silver [13]. Finally, *a direct contrast technique developed by Kiritani and co-workers also has given the nature of* electron-irradiation induced SFT in silver (as well as SFT produced in a variety of materials by quenching, plastic deformation, and by electron, neutron and ion irradiations) as vacancy [14]. The initial erroneous result indicates that nature determinations by the $2^1/_2$D method should be treated with great caution.

The occurrence of vacancy SFT near dislocations in silver is a consequence of the preferential absorption of interstitials due to the dislocation "bias". If the dislocation acts as a good sink, local denudation of interstitials leads to conditions which favour vacancy-cluster formation in the early stages of irradiation. SFT are not seen to have formed in large numbers near irradiated screw dislocations [7], which is as expected for the weak vacancy-screw dislocation interaction.

5. Fe-Ni-Cr ALLOYS

We report here studies of three ternary Fe-x%Ni-17%Cr alloys with Ni content x = 15%, 25% or 40%, and a quaternary alloy containing 15%Ni, 17%Cr and 1%Si. Preliminary accounts of observations in some of these materials are given in references [15] and [16]. Irradiations were performed at temperatures in the range 400°-430°C: At lower irradiation temperatures, little change was observed at dislocations before fine-scale bulk damage developed and quickly masked dislocation images; at higher temperatures, the onset of what may have been σ-phase transformation at the foil surfaces led to a rapid degradation of image quality.

5.1 TERNARY ALLOYS

The observations in the three ternary alloys were qualitatively similar to those made in silver in that lines of cluster damage developed within the compressional strain fields of all dislocations which had a significant edge component. Some of the clusters could be identified as SFT and all are presumed to be vacancy in nature. Dislocations remained dissociated, although dissociation widths decreased. Interstitial climb occurred at favourable sites such as pre-existing jogs. This led to the removal of constrictions along edge and mixed dislocations, following which no further climb was observed. Climb of jogs on screw dislocations produced zig-zag configurations which developed into flat helices. In no case was evidence found for the nucleation of loops directly onto partial dislocations so again the Cu-Al mechanism does not seem operative.

An experiment illustrating some of these observations is shown in figure 3. The material here is Fe-15%Ni-17%Cr, which has a partial separation similar to silver, and the dislocation orientation is about 75°. Before irradiation (figure 3*(a)*) the dislocation is evenly dissociated with no constrictions. After irradiation to a dose of ~ 0.05 dpa at 400°C, an image taken in the same reflection (figure 3*(b)*) shows the partial separation to have narrowed although the dislocation is still dissociated. No constrictions have formed and there is no evidence for any climb of the line. In figure 3*(c)*, in which the partial $\overrightarrow{A\delta}$ is out of contrast, a line of fine-scale cluster damage along the dislocation is evident. This cluster damage lies close to the core of the dislocation. In full contrast experiments neither these clusters nor the larger cluster labelled '*A*' in figure 3*(b)* showed contrast

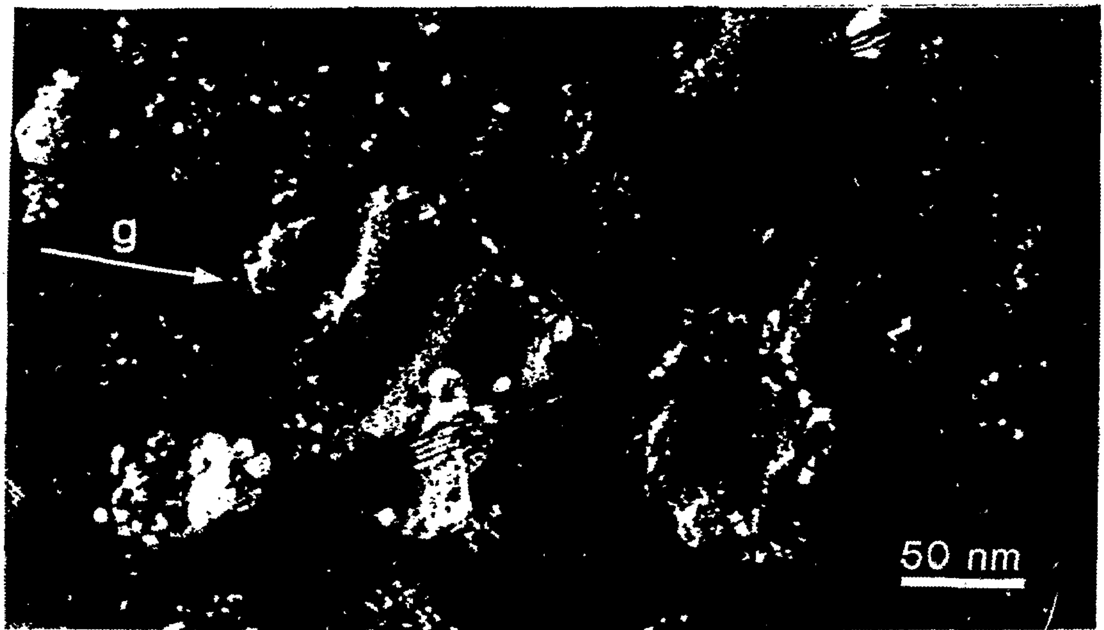

Figure 2: Vacancy SFT nucleated at the inner peripheries of interstitial Frank loops in silver imaged in $\bar g = [1\bar1\bar1]$ with beam direction ($\bar B$) close to $[\bar2\bar1\bar1]$. The Frank loops were formed during an initial irradiation to 1.4×10^{26} e/m^2 at 170°C and the SFT formed subsequently during a room temperature re-irradiation to 6.8×10^{25} e/m^2.

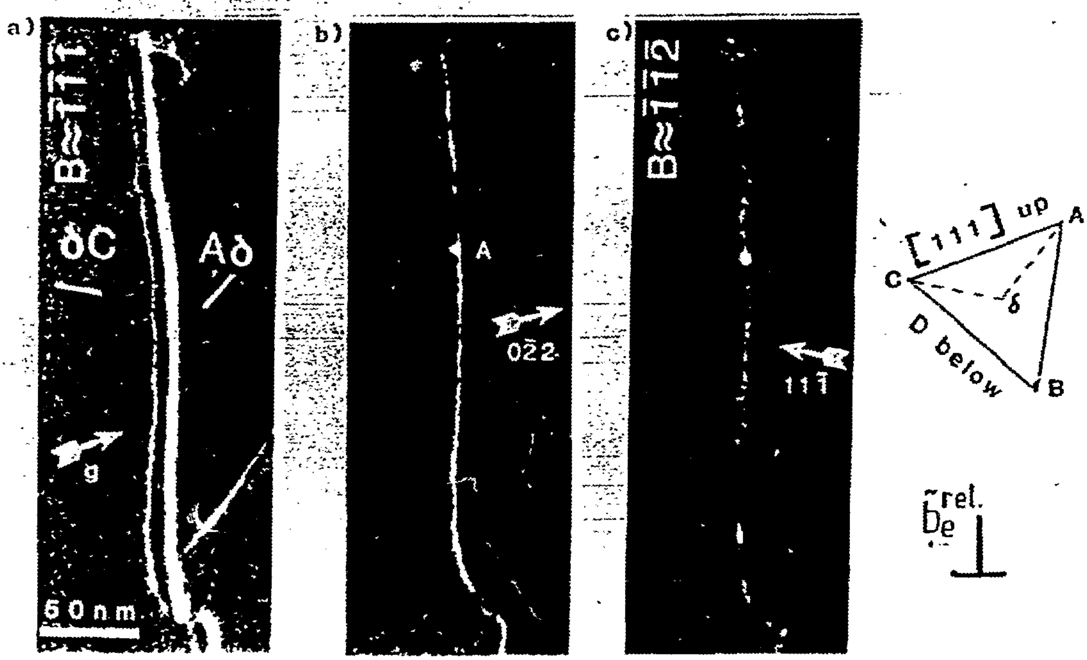

Figure 3: 75° Dislocation in Fe-15%Ni-17%Cr; (a) before irradiation; (b) and (c) after irradiation to 0.05 dpa at ~ 400°C.

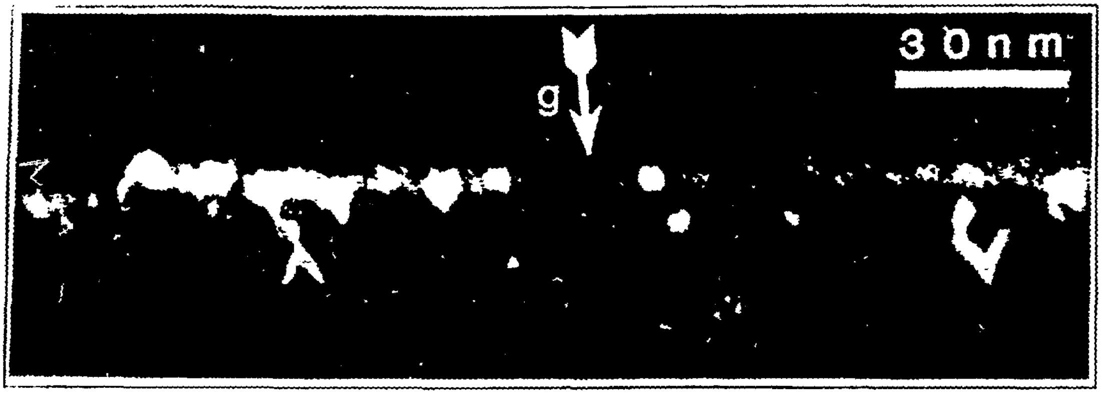

Figure 4: 65° Dislocation in Fe-25%Ni-17%Cr after irradiation to 0.12 dpa at ~430°C. Beam direction ($\vec{B}$) $\approx [\bar{2}\bar{1}\bar{1}]$; $\vec{g} = [\bar{1}\bar{1}1]$.

consistent with any prismatic or Frank loop variant; rather the contrast was consistent with three-dimensional clusters such as SFT. A clearer observation of the nature of these clusters formed along dislocations is provided by figure 4, which shows a dislocation in Fe-25%Ni-17%Cr irradiated to a somewhat higher dose, where it is confirmed that the defects are indeed SFT lying to the compressional side of the dislocation core.

In figure 5 *(a)* the dislocation is initially heavily jogged. Again the material is Fe-15%Ni-17%Cr and the irradiation was to a dose of ~0.05dpa at 400°C. Note that after irradiation (figure 5 *(b)*) the partial separation has decreased and the constrictions have disappeared. Other reflections, not shown, show nucleation of a line of clusters along the dislocation as in figures 3 and 4.

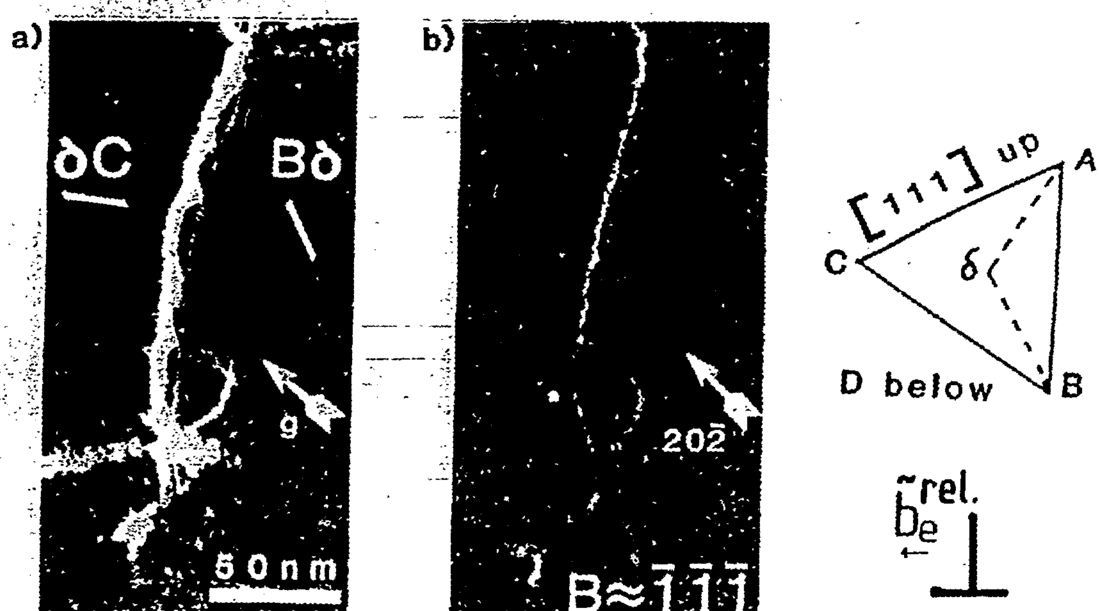

Figure 5: 50-70° Dislocation in Fe-15%Ni-17%Cr; (a) before irradiation; (b) after irradiation to 0.05 dpa at ~400°C.

The screw dislocation of figure 6 is constricted at several points (e.g. J) prior to irradiation (figure 6 *(a)*). After irradiation (figures 6 *(b,c)*) the dislocation has assumed a zig-zag configuration.

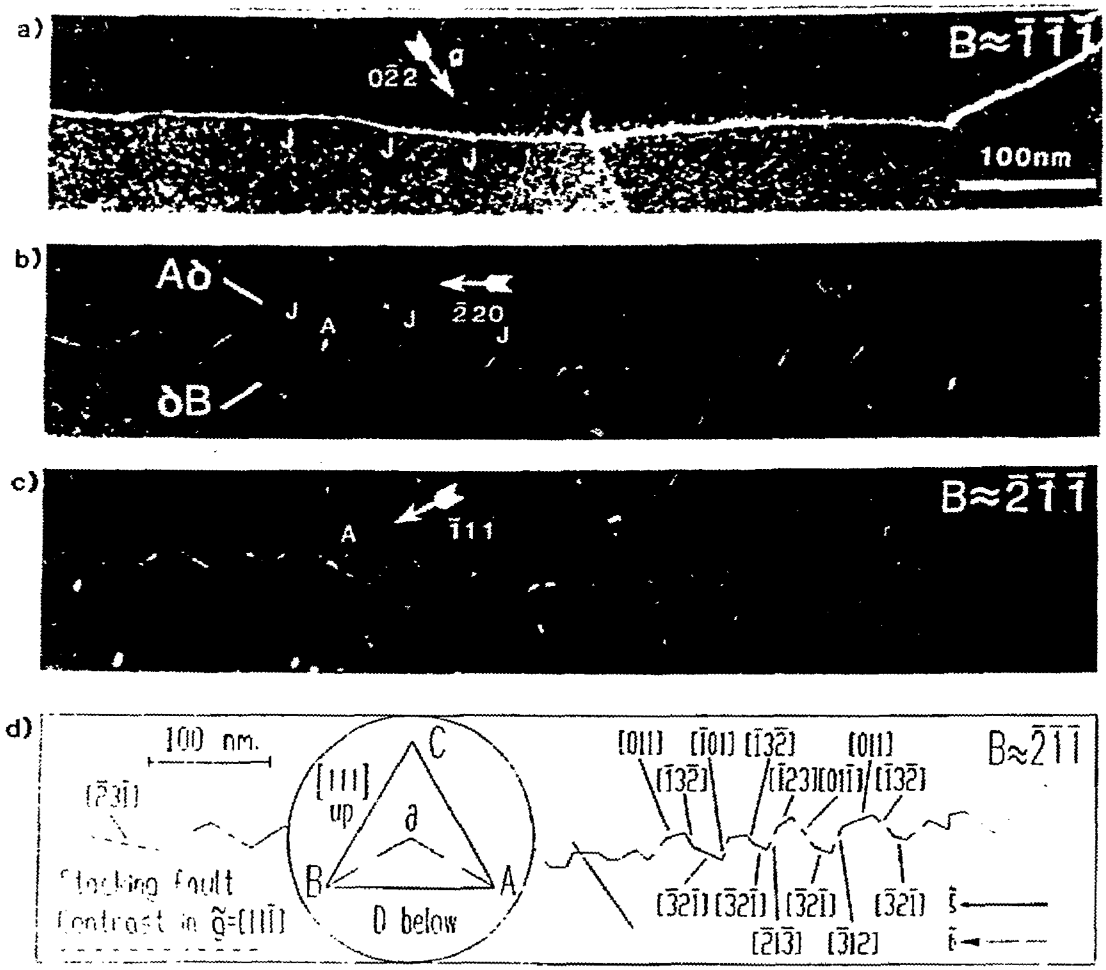

Figure 6: Screw Dislocation in Fe-15%Ni-17%Cr; (a) before irradiation showing positions of pre-existing jogs; (b) and (c) after irradiation to 0.05 dpa at ~400°C; (d) schematic diagram of post-irradiation configuration indicating the line directions of helical segments.

It is possible to correlate some points of the zig-zags with pre-existing jogs, suggesting that climb of these jogs was an essential factor in determining the final configuration. This suggestion is apparently reinforced by the screw dislocation of figure 7, along which no jogs are visible in any reflection taken before irradiation (e.g., figure 7*(a)*) and which has not assumed a zig-zag configuration after irradiation (figure 7*(b)*). In figure 7, note that the large faulted region visible prior to irradiation, the nature of which seems likely to be intrinsic, has been largely annihilated after irradiation.

For a screw dislocation, the jog climb motion is normal to the dislocation line direction and jogs of opposite sign will climb in opposite senses leading to zig-zagging of the line. This is represented schematically in figure 8*(c)*. A careful tilting experiment showed that the dislocation of figure 6 in fact has a flat helical structure after irradiation, still lying essentially in $\delta = (111)$ but with short inclined segments on other {111} planes. The segments appear to align along <110> and <321> directions as represented in figure 6*(d)*. Those segments on δ remain dissociated, whilst the inclined connecting segments appear constricted. The sense of the helix corresponds to net interstitial absorption. Full details of this and other observations of segmented helices will be given elsewhere [17]. The loops in bulk material are interstitial in nature; that labelled '*A*' in figure 6 ($\tilde{b}_L = \alpha\vec{A}$) lies some distance below the dislocation and has not interacted with it. No evidence was found either for the formation of lines of clusters or for any loop nucleation along the screw dislocations of figures 6 and 7.

Finally, we have not attempted to make any distinction above between alloys with different Ni contents. Differences between the three alloys were small. There may be a tendency for vacancy clusters to nucleate at greater distances from the dislocation line and to grow to larger sizes with increasing Ni content [17]. Overall however the behaviour of the ternary alloys was very similar.

5.2 QUATERNARY ALLOY

Observations in the Fe-15%Ni-17%Cr-1%Si alloy were generally rather similar to those made in the ternary alloys. Again partial separations narrowed and

Figure 7: Screw Dislocation in Fe-40%Ni-17%Cr; (a) before irradiation; (b) after irradiation to 0.06 dpa at ~400°C.

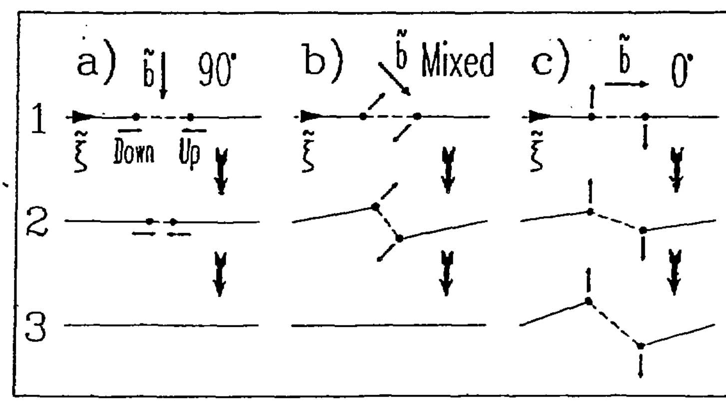

Figure 8: Schematic representation of climb forces acting on jogs under an interstitial super-saturation: (a), (b) and (c) - Edge, mixed and screw dislocations; 1, 2 and 3 indicate initial, proposed intermediate and final configurations. Equal and opposite jogs (labelled "Up" and "Down") experience climb forces in the direction of the small arrows. The Burgers vector $\tilde{b}$ is indicated by the large arrow whilst $\tilde{\xi}$ refers to the dislocation line direction.

lines of vacancy cluster damage developed along the dislocations with appreciable edge components. Screw dislocations assumed zig-zag configurations. An observation not made in the ternary alloys, however, was that new jogs appeared on previously jog-free dislocations, as shown in figure 9, in which an apparently unjogged screw dislocation before irradiation (figure 9(a)) has assumed a zig-zag configuration after irradiation (figure 9(b)) which we believe to represent an early stage in the formation of a segmented helix. For mixed and edge dislocations in this material there were indications that climb by the Thomson & Balluffi mechanism [18] may be occurring [17].

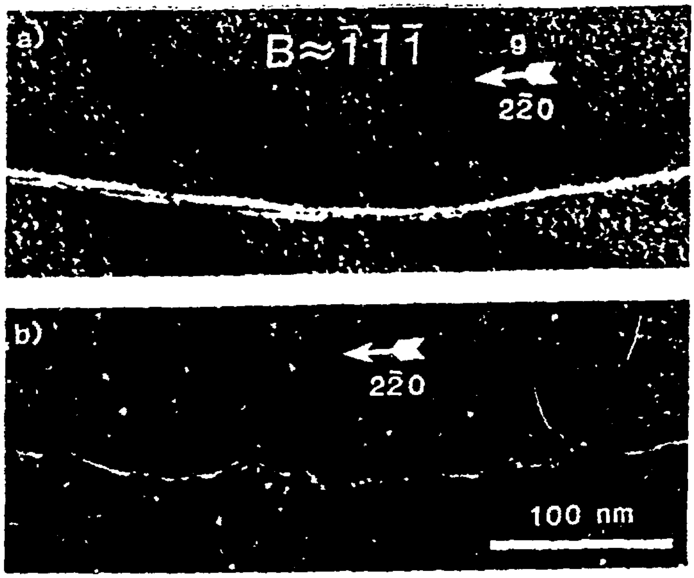

Figure 9: Near-screw Dislocation in Fe-15%Ni-17%Cr-1%Si; (a) before irradiation; (b) after irradiation to 0.09 dpa at ~ 400°C.

6. DISCUSSION

The removal of constrictions along mixed and edge dislocations in the ternary alloys after irradiation (e.g. figure 5) is believed to result from the climbing together, and subsequent annihilation, of pre-existing jogs. As distinct from Cu-Al (figure 1) there is no copious interstitial loop nucleation onto the partials or evidence for the formation of reaction products such as Shockley dipoles. The behaviour of irradiated dislocations in Fe-Ni-Cr is broadly similar to that seen in silver in that dislocations promote the formation of vacancy SFT in their vicinity (e.g., figure 4). The narrowing of partial separations in both Fe-Ni-Cr and Ag may be due to relaxation of the dislocation strain field by these vacancy clusters [9, 16, 19].

Given the high density of vacancy clusters close to dislocations the fate of the more mobile interstitials generated during irradiation must be considered. A greater number of interstitials than vacancies are expected to migrate to dislocations [3,4]. However, the lack of evidence for climb of mixed and edge dislocations in the ternary alloys, or for the nucleation of interstitial loops along dislocations, may suggest that irradiation-generated interstitials are core (or "pipe") diffusing after the annihilation of pre-existing jogs. These jogs are expected to act as barriers to core diffusion [e.g. 20]. The shrinkage of the faulted region connected to the unjogged screw dislocation of figure 7 appears analogous to the shrinkage of "connected voids" on annealing observed by Volin et al [21] but is believed here to indicate interstitial core diffusion under irradiation. Another faulted region (not shown), also in Fe-40%Ni-17%Cr but attached to a heavily jogged dislocation, had shrunk to a much lesser extent after a similar irradiation [17, 22].

The addition of 1%Si to the 15%Ni alloy (figure 9), the only material in which mixed, or edge, dislocations were found to have climbed, is believed to inhibit interstitial core diffusion such that new jogs are created. Silicon is known to migrate towards defect sinks under irradiation [23-25] and, in the case of dislocations, may relax the core to allow the existence of an interstitial super-saturation sufficient to prompt the formation of new jogs. Solute redistribution around the dislocation core (e.g., Hirth & Lothe [26] show the case for oversize solutes in their figures 14-13, 18-4 & 18-9) may similarly inhibit pipe diffusion.

When compared with the ternary alloys, only low vacancy cluster densities are found near climbed dislocations in Fe-Ni-Cr-Si. Vacancy cluster damage is not evident after HVEM irradiation in Cu-13%Al although dislocations in both materials unquestionably act as sinks for vacancies [17, 27]. As alluded to by Cherns [28], the nucleation of interstitial loops directly onto dislocations in the low SFE Cu-13%Al alloy will require the build-up of a sufficient interstitial super-saturation; it would seem plausible, therefore, that dislocations in both CuAl and Fe-Ni-Cr-Si act efficiently as recombination centres, with excess interstitials being absorbed by climb.

By the above model, the development of an interstitial super-saturation is not expected to occur if defects are able to diffuse rapidly to surfaces along dislocations: We therefore suppose that interstitial core diffusion is inefficient along the widely extended dislocations of Cu-13%Al.

7. CONCLUSIONS

1) Dislocations in silver and the ternary Fe-Ni-Cr alloys studied act as good sinks for interstitials, allowing the formation of vacancy cluster damage in their vicinities. Vacancy clusters are seen to form within the compressional strain field of all dislocations which have a significant edge component in these materials.

2) For silver and the ternary Fe-Ni-Cr alloys, interstitials which arrive at dislocations pipe diffuse until they encounter a site, such as a jog, at which they can easily be assimilated. For screw, and near screw dislocations, pre-existing jogs climb apart to form a helical configuration. For edge and mixed dislocations pre-existing jogs may be annihilated. Interstitials now no longer have favourable sites for precipitation and pipe diffuse without further interaction.

3) The addition of silicon to a Fe-Ni-Cr alloy is believed to inhibit pipe diffusion, allowing new jogs to form and enhancing the action of dislocations as recombination centres.

4) The nucleation of interstitial loops directly onto dislocations in Cu-13%Al suggests that pipe diffusion is inefficient in this material.

5) Knowledge of the efficiency of interstitial core diffusion is central to an explanation for microstructural observations.

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