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WEAK-BEAM IMAGING OF DISSOCIATED DISLOCATIONS IN HVEM-IRRADIATED Fe-Ni-Cr ALLOYS*

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Abstract:

We report here on studies by weak-beam electron microscopy of the evolution of microstructures at and near pre-existing line dislocations in a number of Fe-Ni-Cr alloys under electron-irradiation in a high-voltage electron microscope (HVEM). The detailed observations are discussed in terms of dislocation climb mechanisms in these materials and a model based on interstitial pipe diffusion.

1.Introduction

In order for an irradiated material to undergo void swelling, the interstitials and vacancies generated by the irradiation must be mobile and should not migrate in equal numbers to each of the different types of point-defect sink. As early as 1959 [1], it was recognised that a tendency for dislocations to show a preference for attracting interstitials over vacancies may play a crucial role in separating the defect fluxes, thereby allowing excess vacancies to cluster. This preference has been formalized in terms of a dislocation `bias' for interstitials (e.g.[2]) and arises because interstitials have a larger strain field than vacancies and so have a bigger elastic interaction with line dislocations. The objective of the present study was to identify ways in which HVEM-generated point-defects interact with pre-existing line dislocations in Fe-Ni-Cr alloys, with base compositions similar to the stainless steels used in reactors. It was hoped that by studying these interactions, light would be shed on the sink behaviour of dislocations in these materials, possibly leading to a better understanding of their importance to the phenomenon of void swelling.

Previous, similar studies, conducted over the past 12 years in Oxford, Harwell and Argonne have highlighted the complexity of point-defect interactions with dissociated dislocations. In electron-irradiated Cu-13%Al, Cherns et al [3], employing the weak-beam technique because of the fine-scale of damage formed close to irradiated dislocations, were able to deduce that climb occurs via the nucleation of interstitial perfect prismatic dislocation loops (or Frank loops which subsequently unfault) directly onto the individual partials (see, e.g. [4]; Fig. 1); the partial of greater edge character is generally favoured and nucleated loops do not necessarily have the same Burgers vector as the parent dislocation. This results in complex final configurations which frequently contain a number of Shockley dipoles. 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 (see [5]). Importantly, in none of these cases of climb under electron irradiation was any vacancy component of the damage identified, although dislocations unquestionably act as sinks for vacancies (e.g. [6]).

In silver, electron-irradiated at room temperature, however, dislocations were seen to constrict and promote in their vicinity the formation of dislocation loops and (especially) stacking-fault tetrahedra (SFT) [7]. There was no indication of major climb motion of the line, nor of complex interactions between the clusters and line dislocations. The initial confusion regarding the nature of these vacancy SFT formed near dislocations now appears resolved (e.g. [4]). The occurrence of vacancy SFT near dislocations in silver was interpreted as 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 [8], which is as expected for the weak vacancy-screw dislocation interaction.

2. Experimental

The experiments described here were conducted as follows. Electrochemically-thinned single-crystal foils of the chosen alloys (see section 3 below) with foil normals closely parallel to <111> and containing a moderate density of line dislocations were first examined in a conventional transmission electron microscope operated at 200kV. Throughout this study [9], it was found that the dislocation density in the as-received specimens was often ideal, although dislocations were occasionally introduced by slight bending (see [10]). Line dislocations lying in the plane of the foil were imaged using the weak-beam technique, employing the $\{2\bar{2}0\}$ diffraction vectors available at this pole. The Burgers vectors $\underline{b}_p$ of the individual partial dislocations were determined by use of the Howie and Whelan $\underline{g}.\underline{b}=0$ 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 1MeV electrons. 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 [11]. 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.

3. Results

We report here studies of three ternary Fe-x%Ni-17%Cr alloys with Ni content x=15%, 25% or 40%, and two quaternary alloys containing 15%Ni, 17%Cr and 1%Si and 15%Ni, 17%Cr and 2%Mo. Preliminary accounts of observations in some of these materials are given in references [4], [10] and [12]. 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 sigma-phase transformation at the foil surfaces led to a rapid degradation of image quality.

3.1 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 thought to be vacancy in nature. Dislocations remained dissociated, although dissociation widths were often seen to have decreased. Interstitial climb occurred at favourable sites such as pre-existing jogs, leading to the removal of constrictions by mutual annihilation along edge and mixed dislocations, following which no further climb was observed. For screw dislocations, the climb of pre-existing jogs, which is in a direction normal to the dislocation line direction (e.g. [10]), led to zig-zag configurations which further developed into segmented helices. As distinct from Cu-Al, there was no copious interstitial loop nucleation onto the partials or evidence for the formation of reaction products such as Shockley dipoles. These general observations are now illustrated by examples.

Figure 1 shows a 60° dislocation in Fe-25%Ni-17%Cr. Figure 1(a) is taken before, and 1(b) and 1(c) after, irradiation to 0.11 dpa at ~430°C. As is shown by the $\underline{g}.\underline{b}=2$ image in Fig.1a this dislocation contains a large number of constrictions along its length before irradiation, which are likely to indicate the presence of jogs [13]. This dislocation is intersected by two others at a complex node.

After irradiation, almost all of the pre-existing constrictions have disappeared (see also Fig.5 of ref. [4]). This is particularly evident to the left of the node where an even dissociation can be seen in the $\underline{g}.\underline{b}=2$ image of Fig.1(b). In figure 1(c) several small clusters very close to the line are visible. On tilting to the (211) pole (not shown) these clusters were seen to be displaced from the dislocation image in a sense consistent with their lying within the compressional strain field of the dislocation. Several of these clusters, such as that indicated (`T'), show `arrow-head' contrast typical of vacancy SFT.

The screw dislocation of figure 2 is constricted at several points (e.g. `J') prior to irradiation (Fig.2(a)). After irradiation (Figs.2(b,c)), the dislocation has assumed a zig-zag configuration. 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 reinforced by the observations of figure 3, which shows a screw dislocation along which no jogs are visible in any reflection taken before irradiation (such as Fig.3(a)) and which has not assumed a zig-zag configuration after irradiation (Fig.3(b)). In figure 3, note also that the large faulted region visible prior to irradiation, the nature of which seems likely to be intrinsic, has been largely annihilated after irradiation.

A careful tilting experiment, employing reflections in (111), {211} and {011} poles showed that the dislocation of figure 2 in fact has a flat helical structure after irradiation, still lying essentially in δ=(111) but with short inclined segments on other {111} planes. The segments appear to align along <110> and <321> directions as represented schematically in figure 2(d). In common with all other cases of (segmented) helix formation in this study [9], the sense of the helix corresponds to net interstitial absorption. The loops in bulk material are interstitial in nature; that labelled `A' in figure 2 which has $\underline{b}$= αA lies some distance below the dislocation and has not interacted with it. No evidence was found either for vacancy preciptation or for any loop nucleation along the screw dislocations of figures 2 and 3.

Finally, differences between the three ternary alloys were small, although there may have been a tendency for vacancy clusters to nucleate at greater distances from the dislocation line, and to grow to larger sizes, with increasing Ni content (c.f., Fig.3 of reference [4] for the 15%Ni alloy, Fig.1 of this paper for the 25%Ni alloy and Fig.5.2 of reference [9] for the 40%Ni alloy).

3.2. Quaternary alloys

A 60° dislocation in Fe-Ni-Cr-Mo is shown in figure 4. Before irradiation (Fig.4(a)), this dislocation possessed a complex structure, and evidently contained a large number of constrictions. A particularly sharp cusp may indicate pinning of the line. As in the case of the ternary alloys, irradiation removed these constrictions, to leave the dislocation evenly dissociated (confirmed by $\underline{g}.\underline{b}$=2 images, not shown). Dissociated, straight line sections have not apparently climbed and the Burgers vector of the dislocation is unchanged. Vacancy clusters have formed close to the line but no loops have nucleated directly onto partials. The dislocation has also straightened and the large, fully-formed, pre-existing SFT at left has been partially eroded.

Irradiated dislocations in Fe-Ni-Cr-Si also behaved in a manner similar to dislocations in the other materials. Screw dislocations assumed zig-zag configurations [4], partial separations were again often seen to have narrowed and vacancy cluster damage developed along dislocations with appreciable edge components. Also, in common with Fe-Ni-Cr-Mo, dislocations often possessed a complex structure before irradiation. As distinct from the other alloys, however, vacancy cluster densities close to dislocations frequently were low, dislocations were commonly found to be heavily constricted after irradiation, and new jogs were found on previously jog-free dislocations [4,9].

Some of these observations are shown in figure 5, in which a heavily constricted, near-60° dislocation, which is apparently pinned prior to irradiation (Fig.5(a)), has a large number of new super-jogs (e.g. `J') along its length after irradiation (Figs. 5(b) to 5(d)). Note the characteristic `offset' contrast exhibited by the super-jogs [14-16], which, employing large angle tilting experiments such as that shown in figures 5(b) to 5(d), were measured to be ~10-15nm high. The proximity of climbing jogs to the P_a node at right may have aided, or brought about, its constriction after irradiation [17]. A generally low density of vacancy clusters, sometimes identifiable as SFT (ringed in Fig.5(c)), also was evident.

4. Discussion

In all of the alloys studied, and also in silver, several factors suggest that irradiation-generated interstitials move easily along dislocations by pipe diffusion. These factors include: the lack of evidence for climb of mixed and edge dislocations in the ternary alloys, or for the nucleation of interstitial loops along dislocations; and the fact that a greater number of interstitials than vacancies is expected to migrate to dislocations, yet the observed clusters are all of vacancy type. These observations can be rationalised if it is assumed that once jogs or other obstacles to pipe diffusion are annihilated, interstitials can move freely along the dislocation to the foil surface. Thus the interstitial supersaturations necessary to nucleate new jogs or loops and initiate climb by the Thomson-Balluffi or Cherns mechanisms never develop. The unjogged screw dislocation of figure 3 which is seemingly unaffected by irradiation, whilst the faulted region at its end has been almost completely annihilated (see also [18]), provides rather direct evidence for this process. Recent molecular dynamics simulations [19] have highlighted the importance of interstitial (as opposed to vacancy) core diffusion. It is interesting to note that the contention that interstitials are more likely to reach jogs by bulk rather than core diffusion (e.g. [20]) is not borne out by the present study.

A common feature in Fe-Ni-Cr-Mo, seen to varying degrees in all our observations of dislocations in this material, is the particularly complex morphology of dislocations prior to irradiation [9]. We believe this to be a result of the thermal segregation of Mo to dislocations during crystal growth and subsequent cooling. The segregation of Mo to and along grain boundaries, which is thought to occur via a bound vacancy complex, has previously been noted both during ageing and, in particular, during slow cooling [21,22]. Under irradiation, however, Mo has been shown to diffuse rapidly away from defect sinks, such as dislocations, via the inverse Kirkendall effect [23-26]. It is possible, therefore, that many of the constrictions seen along dislocations in the Mo-doped alloy prior to irradiation arise not from jogs but from solute pinning, and that the disappearance of these constrictions under irradiation is the result of the reverse segregation of Mo away from, and along, dislocations. The observation that only a few of the pre-existing constrictions along a screw dislocation in Fe-Ni-Cr-Mo acted as base-points for the generation under irradiation of a segmented helix [9] supports this view. This de-segregation, and the fact that the addition of Mo apparently has little effect (e.g. Fig.4) upon the initial irradiation response of dislocations, may suggest that the importance of molybdenum to the phenomenon of void swelling lies principally in defect-trapping. Ashworth, in his study of radiation-induced segregation in Fe-Ni-Cr alloys [23], also advances this hypothesis.

The only material in which edge or mixed dislocations showed evidence of climb once pre-existing jogs had been annihilated was the alloy containing silicon (Fig.5). Thermal segregation of silicon may explain the complex initial configuration of this dislocation. As distinct from Mo, however, silicon is known to migrate towards defect sinks under irradiation [25-27]. A build-up of silicon at a dislocation may sufficiently relax the core that interstitial pipe diffusion is inhibited. In turn, this may allow the development of an interstitial super-saturation sufficient to prompt the formation of new jogs and so lead to climb. Solute redistribution around the dislocation core (e.g., Hirth & Lothe [28] show the case for oversize solutes in their figures 14-13, 18-4 & 18-9) may similarly inhibit pipe diffusion. Since the nucleation of new jogs must require a non-zero interstitial super-saturation (e.g. [5]), the absorption of interstitials at dislocations in Fe-Ni-Cr-Si therefore is nucleation rate-controlled rather than diffusion rate-controlled and dislocations in Fe-Ni-Cr-Si may not be described as perfect sinks for interstitials. The mechanism of climb itself is still uncertain, although the lack of loops with Burgers vectors other than that of the parent dislocation may suggest the Thomson & Balluffi mechanism [29]. One must consider, however, whether loops $\underline{b}$= AB are likely to nucleate in 60° orientation.

The build-up of an interstitial supersaturation, possibly reducing the effective dislocation bias, may also explain the low vacancy cluster densities found near climbed dislocations in Fe-Ni-Cr-Si since dislocations may act efficiently as recombination centres. Finally, it is interesting to note that the clustering of vacancies close to dislocation cores, which must require a non-zero vacancy supersaturation, seemingly contradicts the widely held view that pipe diffusion of vacancies is particularly rapid (e.g. [30]).

5. Conclusions

1) Dislocations in the ternary Fe-Ni-Cr alloys and in the Fe-Ni-Cr-Mo alloy 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 Fe-Ni-Cr-Mo and the ternary Fe-Ni-Cr alloys, interstitials which arrive at dislocations are believed to 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 to the foil surface.

3) The addition of Mo to a Fe-15%Ni-17%Cr alloy apparently has little effect on the irradiation response of dislocations. This may suggest that the origin of any change in swelling response of steels on adding Mo does not lie on its effect upon dislocations.

4) The addition of silicon to a Fe-Ni-Cr alloy has a significant effect upon the irradiation response of dislocations. Segregation of Si to dislocations is believed to inhibit pipe diffusion, thereby allowing new jogs to form and also enhancing the action of dislocations as recombination centres.

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

6. Acknowledgements

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Captions:

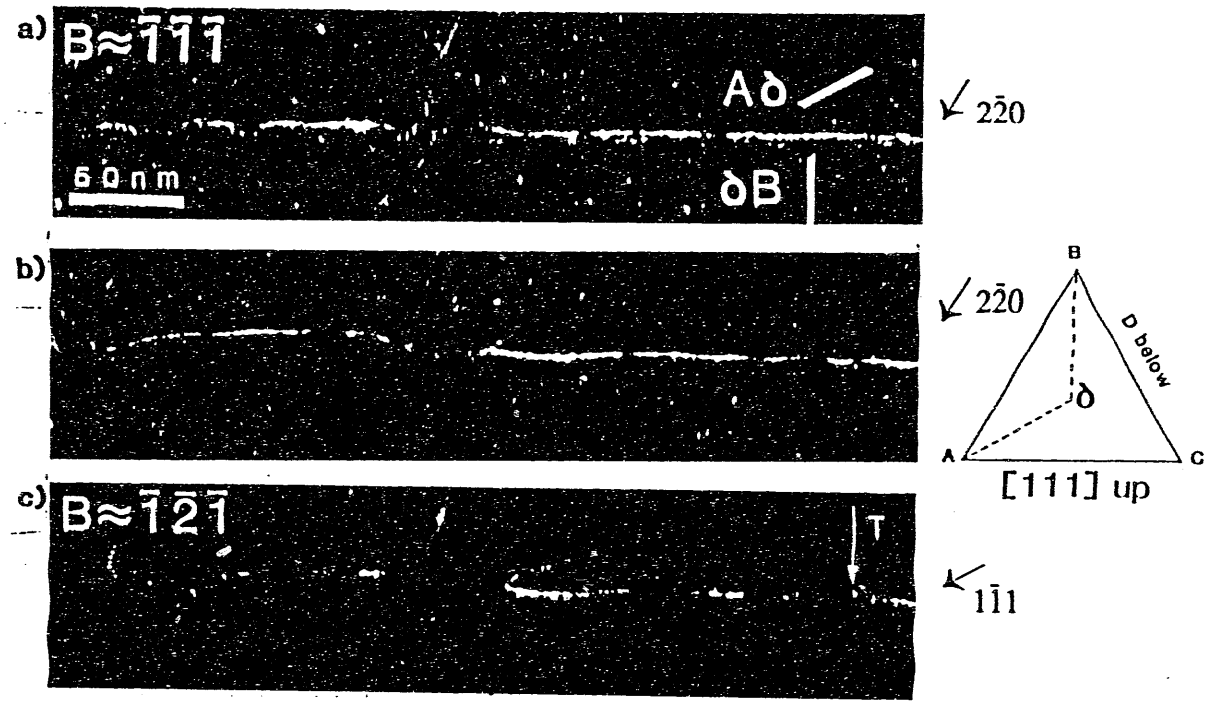
Figure 1: 60° Dislocation in Fe-25%Ni-17%Cr; (a) before irradiation, (b) and (c) after electron irradiation to 0.11 dpa at ~430°C.

Figure 2: Screw dislocation in Fe-15%Ni-17%Cr; (a) before irradiation showing positions of pre-existing jogs; (b) and (c) after electron irradiation to 0.05dpa at ~400°C; (d) schematic diagram of post-irradiation configuration indicating the line directions of helical segments.

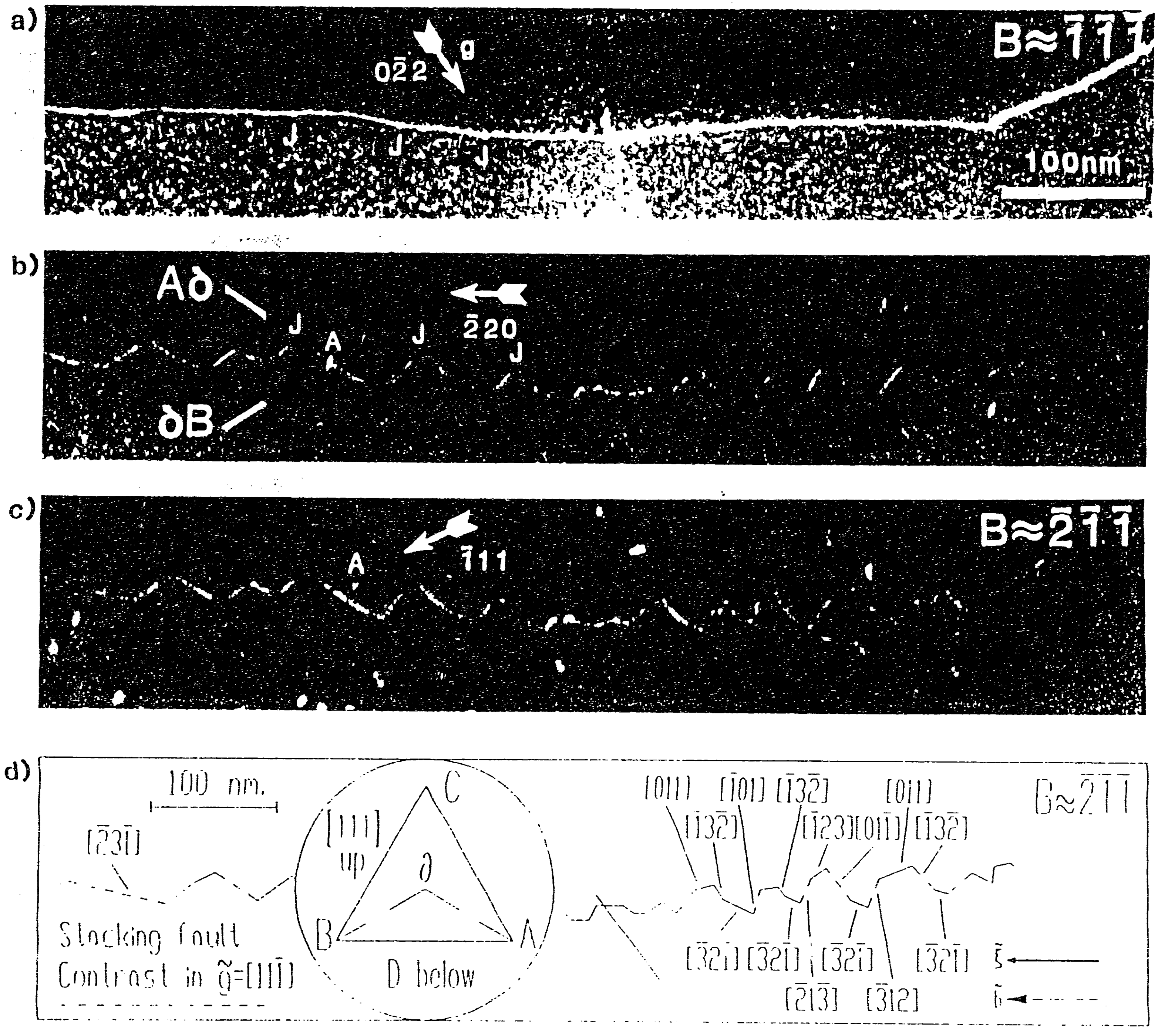
Figure 3: Screw dislocation in Fe-40%Ni-17%Cr; (a) before irradiation; (b) after electron irradiation to 0.06 dpa at ~400°C.

Figure 4: 60° Dislocation in Fe-15%Ni-17%Cr-2%Mo; (a) before irradiation; (b) and (c) after electron irradiation to 0.06 dpa at ~420°C

Figure 5: 60° dislocation in Fe-15%Ni-17%Cr-1%Si; (a) before irradiation; (b) to (d) after electron irradiation to 0.13 dpa at ~400°C.



CAN 92 Si [illegible] ML JENKINS FIGURE 1



a)
0$\bar{2}$2
g
J
J
J
B≈$\bar{1}\bar{1}\bar{1}$
100nm
b)
A∂
∂B
J
A
J
$\bar{2}$20
J
c)
A
$\bar{1}$11
B≈$\bar{2}\bar{1}\bar{1}$
d)
100 nm.
[$\bar{2}$3$\bar{1}$]
Stacking fault
Contrast in $\tilde{g}$=[11$\bar{1}$]
[111]
up
C
∂
B
A
D below
[011]
[$\bar{1}$01]
[$\bar{1}$3$\bar{2}$]
[011]
[$\bar{1}$3$\bar{2}$]
[$\bar{1}$23]
[01$\bar{1}$]
[$\bar{1}$3$\bar{2}$]
[$\bar{3}$2$\bar{1}$]
[$\bar{3}$2$\bar{1}$]
[$\bar{3}$2$\bar{1}$]
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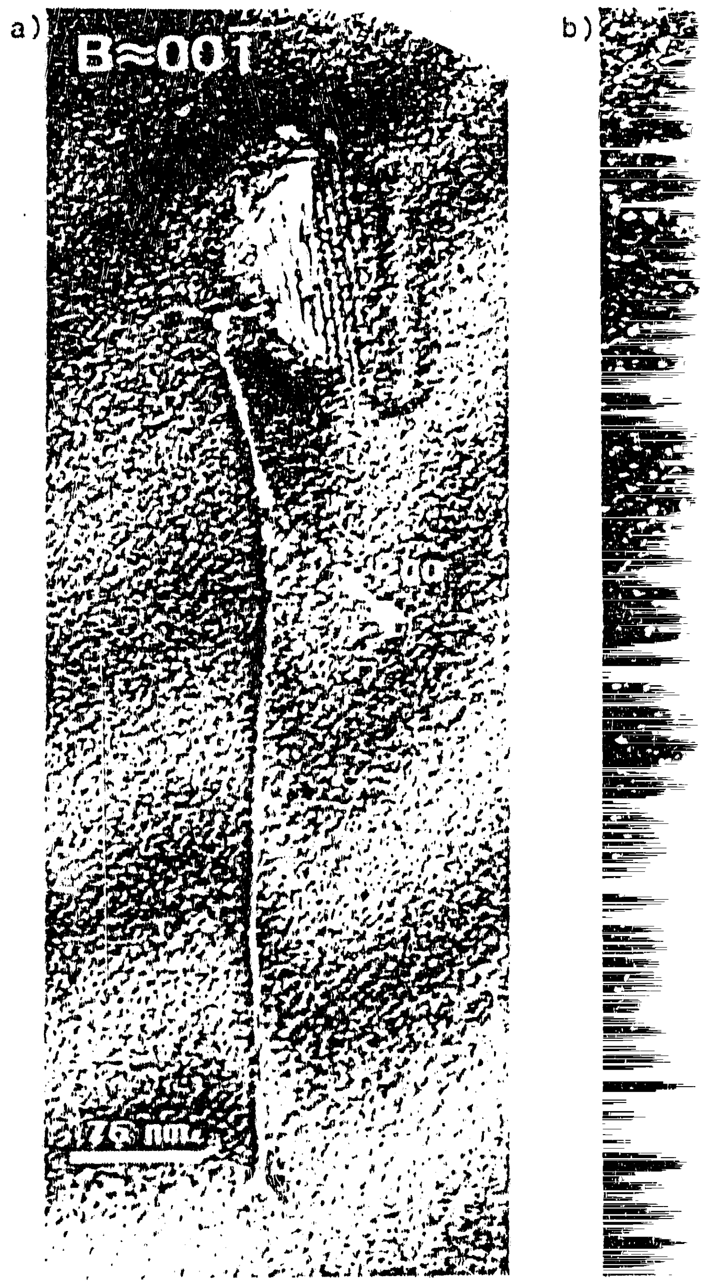
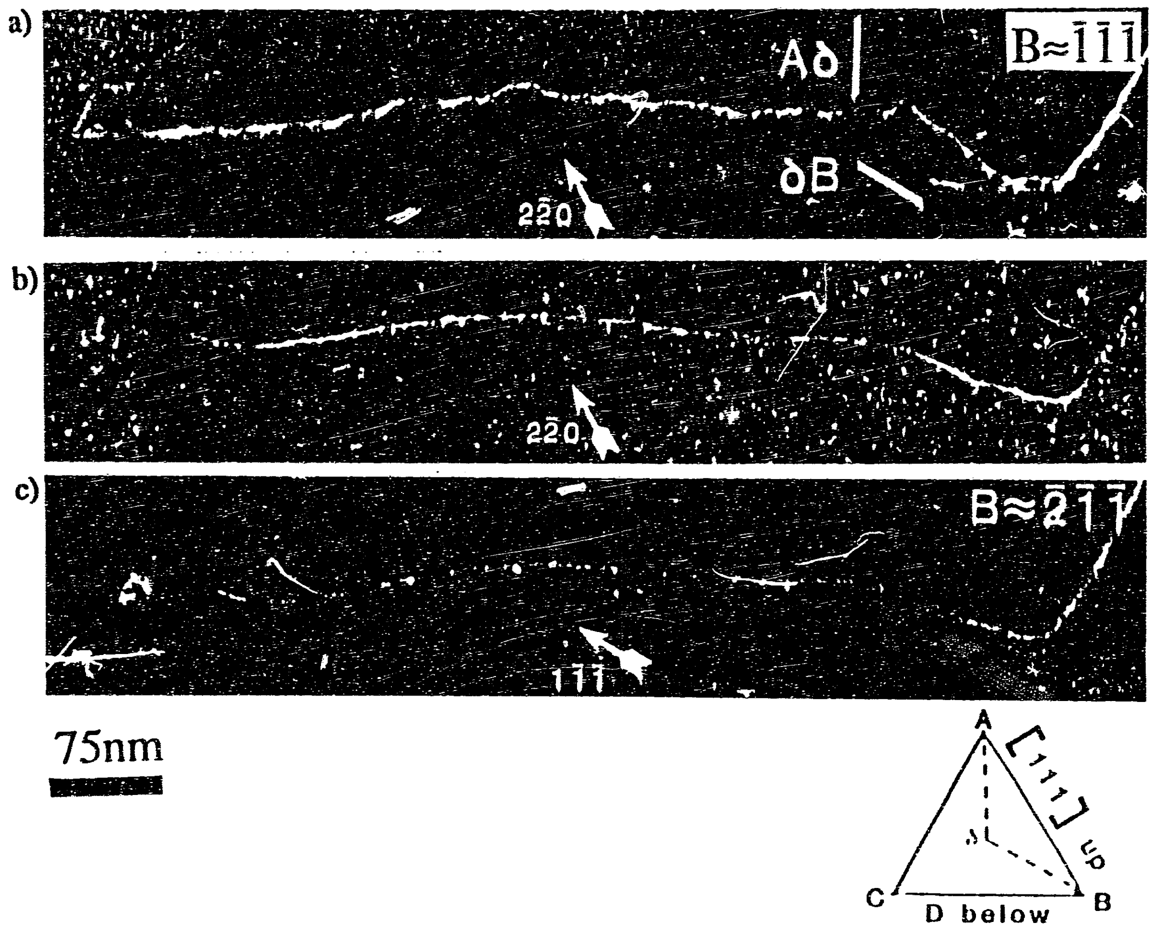
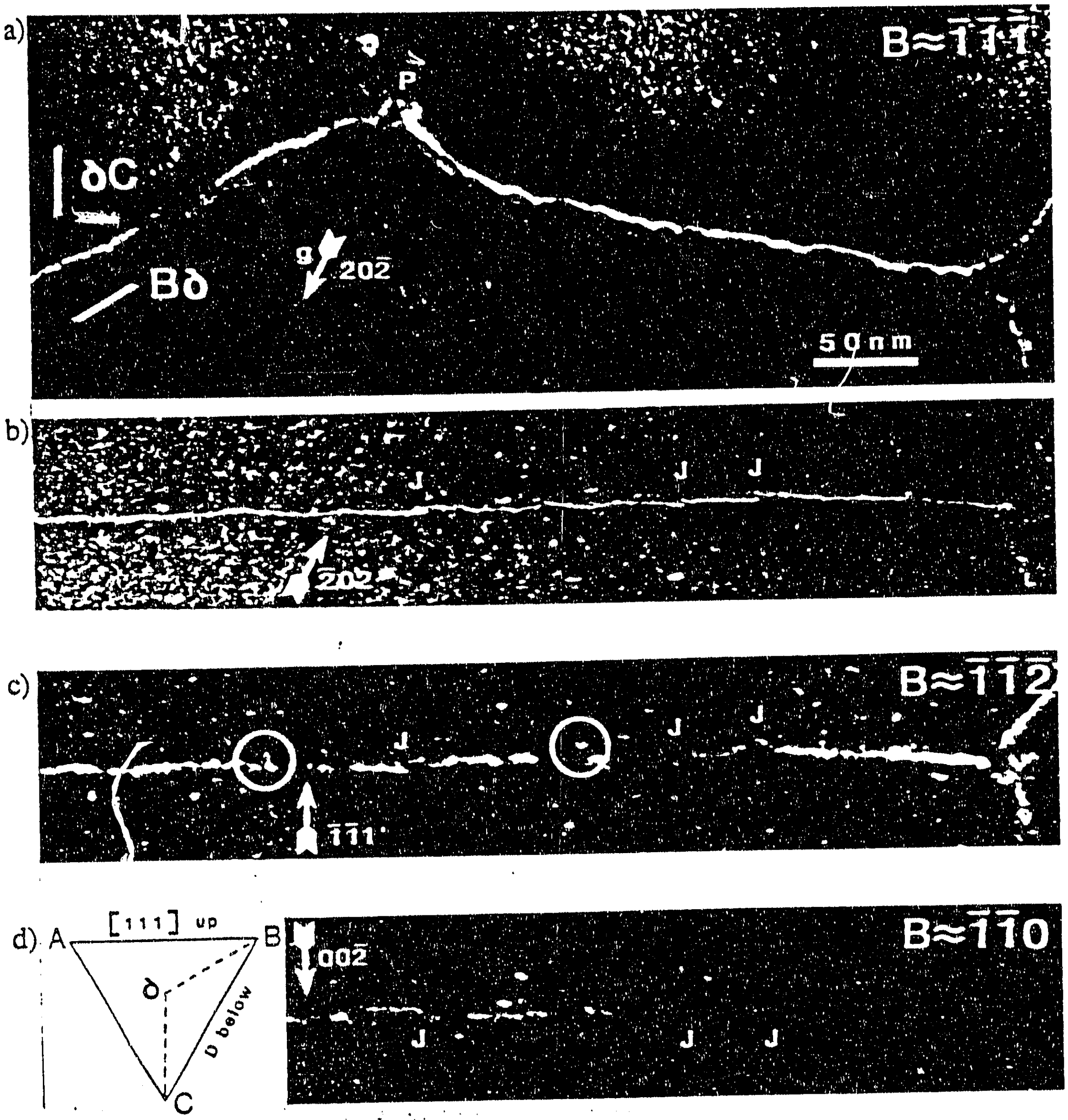
[$\bar{2}$1$\bar{3}$]
[$\bar{3}$12]
B≈$\bar{2}\bar{1}\bar{1}$

3




a)
b)
c)
Aδ
δB
B≈1̄1̄1̄
22̄0
22̄0
B≈2̄1̄1̄
11̄1̄
75nm
A
B
C
[111] up
D below



CAN 92 King, Jenkins FIGURE 5

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