INTERSTITIAL REMOVAL IN STAGE-III RECOVERY OF NEUTRON-IRRADIATED W†

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We have previously indicated by direct observation that an intrinsic defect is responsible for the recovery in stage-III annealing of neutron-irradiated platinum[1] and that this defect is consistent with a ⟨100⟩ split interstitial as has been previously proposed.[2-4] Since irradiated bcc materials exhibit a similar recovery spectrum to that of fcc metals, it is of interest to examine the possibility of a further correspondence between materials of the two structures with respect to the specific defects involved in the specific recovery stages. In particular, some evidence has been given that the defect involved in stage-III recovery in bcc crystals is an extrinsic defect.[5] This evidence is derived from a comparison of the resistivity changes of rather impure materials with somewhat purer materials. What, however, is not precisely known in these experiments is the concentration of defects and therefore what fraction of the defects is pinned by impurities. In the case of tungsten, stage III is present even though the material has a very small amount of impurities in solution[6]; i.e., it is apparently an intrinsic property of the material. Further, one can irradiate the material to exposures which introduce defect concentrations far in excess of the impurity concentration, which should allow one to distinguish an intrinsic recovery process from an extrinsic one. We have undertaken some experiments which directly bear on these questions and are of interest to studies of defects in general.

The technique used to observe the irradiated material directly was a field-ion microscope operated at 4.2°K.[7] The tungsten was obtained from the Westinghouse Corporation as 5-mil wire drawn down from triple-pass zone-refined single-crystal material. The residual resistivity ratio of this starting material was greater than 40 000. This material was irradiated in the Brookhaven High-Flux Beam Reactor to an exposure of 5×10^{19} fast neutrons/cm^2 ($E>1$ MeV) at approximately 100°C and subsequently examined in the field-ion microscope.

Repeated examination of the material below stage-III recovery revealed single vacancies, clusters of vacancies, and single interstitials. The interstitial concentration was approximately 1×10^{-3} at.%. These defects appear as bright spots the size of which are larger than atoms in normal lattice sites. This is consistent with previous observations of interstitials in W and Pt.[8,9] Occasionally, one of these interstitial atoms was seen to appear suddenly at the surface and remain there. This phenomenon has also been observed in tungsten that has been electron irradiated below state-III recovery.[10] Müller has previously suggested that these are interstitial atoms lying a few atomic layers deep which are pulled out to the surface by the field stress.[8] A micrograph of one such interstitial atom is shown in Fig. 1(a), with the position of the interstitial mapped out in

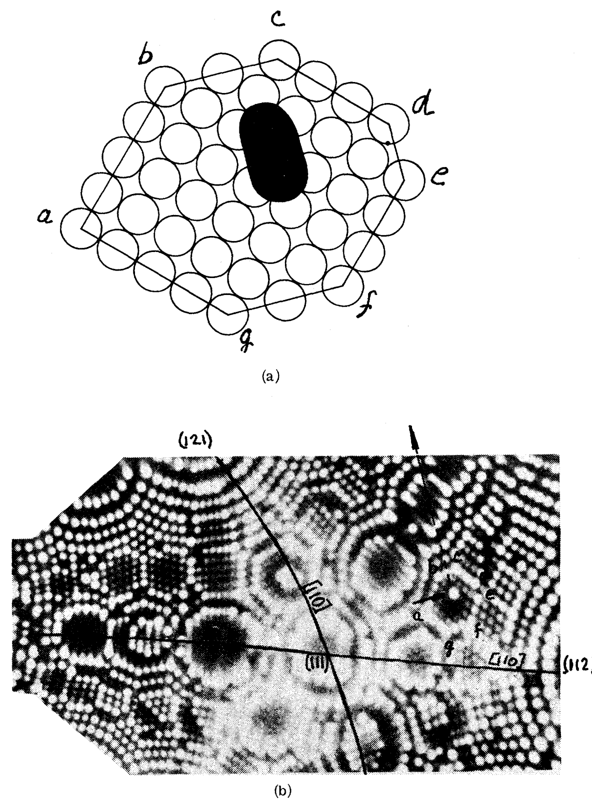

FIG. 1. (a) Interstitial atom in a closed net plane in tungsten irradiated with 2×10^{20} fast neutrons/cm^2 below stage-III recovery. (b) Interstitial shown in (a) mapped out.

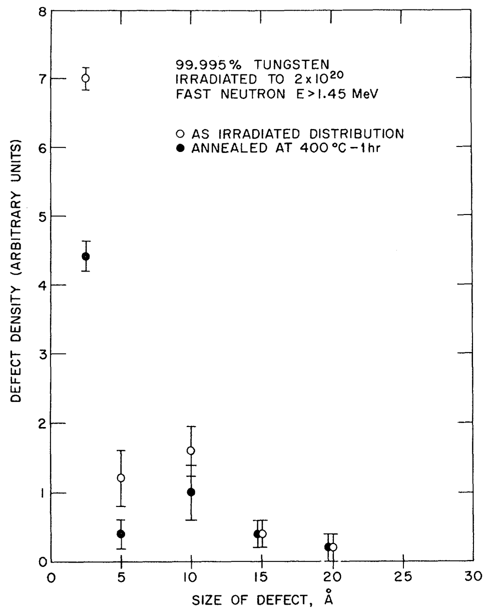

FIG. 2. Vacancy-type defect distribution in tungsten before and after stage-III recovery.

Fig. 1(b). Several characteristic features of these interstitial atoms are illustrated by this micrograph. The positions of the bright spots do not correspond to atoms in ordinary lattice positions either in the plane observed or in the one directly above or below it; furthermore, the size of these bright spots is considerably larger than that of those atoms lying within the plane. These observations indicate that we are in fact "seeing" a displaced atom—an interstitial—lying in a subsurface position. The shape of the bright spots is also seen to be anisotropic, i.e., extended in one direction. When this anisotropy exists, the direction is always consistent with that of a ⟨110⟩ split interstitial in the bcc lattice.

Annealing the material at 400°C for 1 h causes a drop in the concentration of these interstitials from approximately 0.1 at.% to less than 10^{-3} at.%. Simultaneously the vacancy concentration decreases from 0.2 to 0.13 at.%. Changes in the vacancy-type defect size distribution upon annealing at 400°C, for 1 h are shown in Fig. 2. The only possible explanation of a drop in the vacancy concentration from 0.2 to 0.13 at.% upon annealing through stage III is the migration of interstitials to vacancies, since it has previously been shown that in tungsten extensive vacancy motion occurs in the temperature range between 700 and 900°C.[11,12] Thus, the simultaneous decrease in the concentration of interstitials and vacancies can only be accounted for by the migration and subsequent annihilation of the interstitials at vacancy-type defects already present in the irradiated material.

The possibility that the observed interstitial atoms are impurity-trapped interstitials can be discounted since the ratio of interstitials to total impurities present is approximately 20/1 for this material. Thus, one impurity atom must trap more than 20 interstitial atoms to account for 10^{-1} at.% of interstitials. In our observations we observe single interstitials but certainly not large interstitial clusters.

The observations thus indicate that stage-III recovery in neutron-irradiated tungsten is an intrinsic recovery stage in which the removal of a self-interstitial-type defect occurs. The main sinks for these defects are the irradiation-produced single vacancies.

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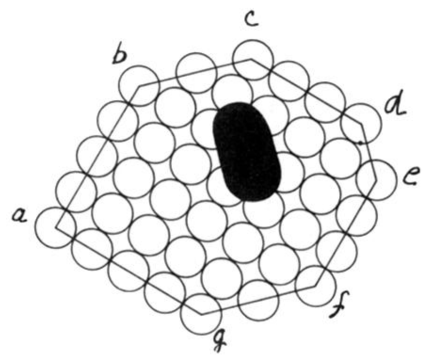
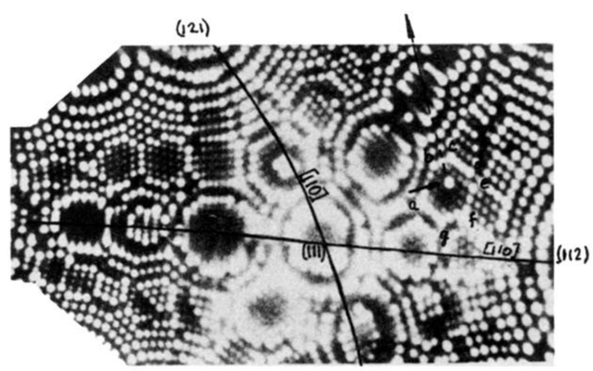


(a)



(b)

FIG. 1. (a) Interstitial atom in a closed net plane in tungsten irradiated with 2×10^{20} fast neutrons/cm^2 below stage-III recovery. (b) Interstitial shown in (a) mapped out.